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CHARACTERIZATION OF THE SOLID,
AIRBORNE MATERIALS CREATED BY THE
INTERACTION OF UF_6 WITH ATMOSPHERIC
MOISTURE IN A CONTAINED VOLUME

P. W. PICKRELL

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SUMMARY

Several experiments have been performed in which UF_6 was released into air under static conditions in a 6 m^3 release chamber in order to characterize the solid products of hydrolysis as the amount of UF_6 released was increased. An aerosol concentration of $\sim 25 \text{ gm/m}^3$ (150 gm) was the largest achieved. Electron microscopy was used to determine the morphology of the aerosol particles and to obtain geometric size measurements from micrographs and a cascaded impactor was used to obtain aerodynamic size measurements by measuring particle masses. Particle sizes and rate of particle size change were observed to be dependent on aerosol concentration.

INTRODUCTION

The work reported herein represents a part of the support being provided to the technical community on the effects of the reactions of UF_6 with atmospheric moisture. This work was originally associated with the UF_6 Release Safety Analysis Report program but is now carried as a part of the Plant and Long Range Technical Support (P&LRTS) environmental program at the Oak Ridge Gaseous Diffusion Plant. The goal of the work is to determine the chemistry and physics of the UF_6 -atmospheric moisture reaction under a variety of conditions so that information about resulting species and product morphologies is available for containment and removal (knockdown) studies as well as for dispersion plume modeling and toxicology studies. This report completes the P&LRTS milestone for reporting the data from releases in the large containment chamber. Experiments to characterize smaller releases have been described in an earlier report.

The Safety Analysis Report (SAR) effort to determine the fate of UF_6 released to the environment is primarily concerned with evaluating and predicting the health hazards associated with UF_6 releases and includes both toxicity data and dispersion modeling of such releases. Information about the character of the particles comprising the aerosol associated with such releases is relevant to both modeling and toxicity. Some experiments to characterize such aerosol particles were described in an earlier report,* and this report summarizes some follow-on experiments conducted with larger amounts of UF_6 in a larger release chamber with the intent of progressively increasing the amount of UF_6 released to see the effect of increasing concentration on particle morphology. As before, only the stable, long-term solid hydrolysis products were examined. Transmission electron microscopy and microbalance measurements were the primary techniques used to characterize the materials described in this report since morphology

*P. W. Pickrell, Characterization of the Solid, Airborne Materials Created by the Interaction of UF_6 with Atmospheric Moisture in a Contained Volume, K/PS-144, Union Carbide Corp. Nuclear Div., Oak Ridge Gaseous Diffusion Plant, April 1982.

(size and shape) was of primary interest in this series of experiments, but X-ray diffraction and wet chemical methods were used for evaluation in some instances.

EXPERIMENTAL

The experiments described in this report were conducted in a release chamber constructed of 3/4 in. thick Plexiglas sheets. The outside dimensions of the chamber are 5 ft x 6 ft x 7 ft and the volume is $\sim 6 \text{ m}^3$. The containers that held the UF_6 to be released were made of nickel tubing of various sizes. In some of the experiments, a dependable method of release was not used, and the release time and completeness were not controllable. Trial and error experience allowed a reliable and effective mechanism for controllable release of the UF_6 to be developed and used for two of these experiments. This consisted of using an oversized Teflon plug with an O-ring seal to cover the end of the tube containing the UF_6 . This plug could be manually removed when desired, thereby eliminating restrictions caused by valves, rupture holes, etc. Samples for transmission electron microscopy were collected on Formvar-coated nickel microscope grids. Efforts to minimize "pile-up" of particles, which would have the effect of preventing observation of the actual airborne entities, consisted of controlling time of collection, when particles were collected by gravitational settling on the grids, or sampling a limited amount of aerosol in one experiment when the aerosol particles were centrifuged onto the microscope grids. Aerodynamic particle size distributions were obtained with a cascaded, quartz-crystal microbalance instrument, aerodynamic size being the diameter of the spheres which the particles would mimic in aerodynamic behavior. Attempts to measure the rate of settling of the aerosol produced in some of these experiments by weighing the settled material were not successful because of erratic behavior of the weighing device or nonuniform deposition of the settled product. Turbulence and considerable charging by static electricity in the release chamber seemed to be the major causes of these problems.

FIRST ONE-GRAM RELEASE

An experiment in which 1 g of UF_6 was to be released into a 200 ft³ (6 m^3) release chamber was only partially successful when localized heating of the UF_6 container caused premature rupture of the container which was a nickel tube being used as the heating element as well as the container. A plug of solid UF_6 developed at the rupture site and only 5% or 50 mg of UF_6 was actually released. Figure 1 is a photograph of the container after the release, showing the small rupture opening. The air temperature in the chamber at the time of release was 20°C and the RH was 75%. The temperature of the wall of the UF_6 container, measured midway between the ends where electric leads were clamped, was 64°C (the melting point of UF_6) at time of release. The thin aerosol produced by this release was dispersed throughout the chamber within 3 min and totally fallen out between 26 and 40 h after release as determined by



FIGURE 1
PHOTOGRAPH OF 1/8" DIA. CONTAINER AFTER RUPTURE AND RELEASE
FIRST 1-GRAM RELEASE

laser light scattering. Electron micrographs of settled aerosol particles show them to be 0.1 to 0.3 μm spheroids with some agglomeration occurring after several hours, but the low particle concentration prevented much or large agglomerate formation. Figures 2 and 3 exhibit the sparsity of aerosol particles and the lack of agglomeration, collection time for Fig. 2 being 1 1/4 h and for Fig. 3 17 h.

SECOND ONE-GRAM RELEASE

The release of 1 g of UF_6 in a nickel tube container when the container wall was at 95°C , i.e., $\sim 30^\circ\text{C}$ above the triple point of UF_6 , produced aerosol particles which appeared to contain unhydrolyzed UF_6 , as well as UO_2F_2 , for some time, perhaps as much as 2 h, after the release occurred. The presence of the UF_6 in the UO_2F_2 matrix was evidenced by the formation of faint halos around some of the particles after collection. These halos or shadows are caused by the continued localized hydrolysis of UF_6 as it emanates from the collected particles. This is the same effect on a smaller scale often seen when particles of UF_6 and UO_2F_2 large enough to be seen by the unaided eye are produced in UF_6 releases. Figure 25 is a photograph of larger particles and associated halos produced by a larger release (35 g) of UF_6 . The container for the 1 g of UF_6 was a straight-walled nickel tube with a rupture diaphragm of 0.001-in. thick nickel butt-soldered onto the end. When the container and contents (the container itself was used as the resistance heating element) reached 95°C , the diaphragm solder joint failed which had the effect of immediately creating an opening as large as the container bore. This allowed more than 95% of the material to be expelled in less than 1 sec, a much shorter time than is required when metal container rupture occurs. Figure 4 is a photograph of the end of the container with the diaphragm still partially attached. The air temperature and relative humidity into which the UF_6 was released were 21°C and 75%, respectively. Table 1 gives the aerodynamic particle size distributions obtained on the aerosol at various elapsed times after the release. The distributions were obtained with a cascaded quartz crystal microbalance impactor. As can be seen, the aerosol particles grow with elapsed time until preferential fallout causes a decrease in aerosol particle size as reported for earlier experiments.* Unlike most earlier experiments, however, the increase in aerosol particle size is not predominantly the result of particle agglomeration but of continued particle growth as evidenced by the electron micrographs shown in Fig. 5 and 6 of samples collected for the first 10 min after release and for 6 h beginning 3 h after the release. This continued growth is attributed to continued UO_2F_2 accretion as the occluded UF_6 continued to hydrolyze. Preferential fallout is considered to be the cause for the decrease in size at longer times.

*Ibid K/PS-144.

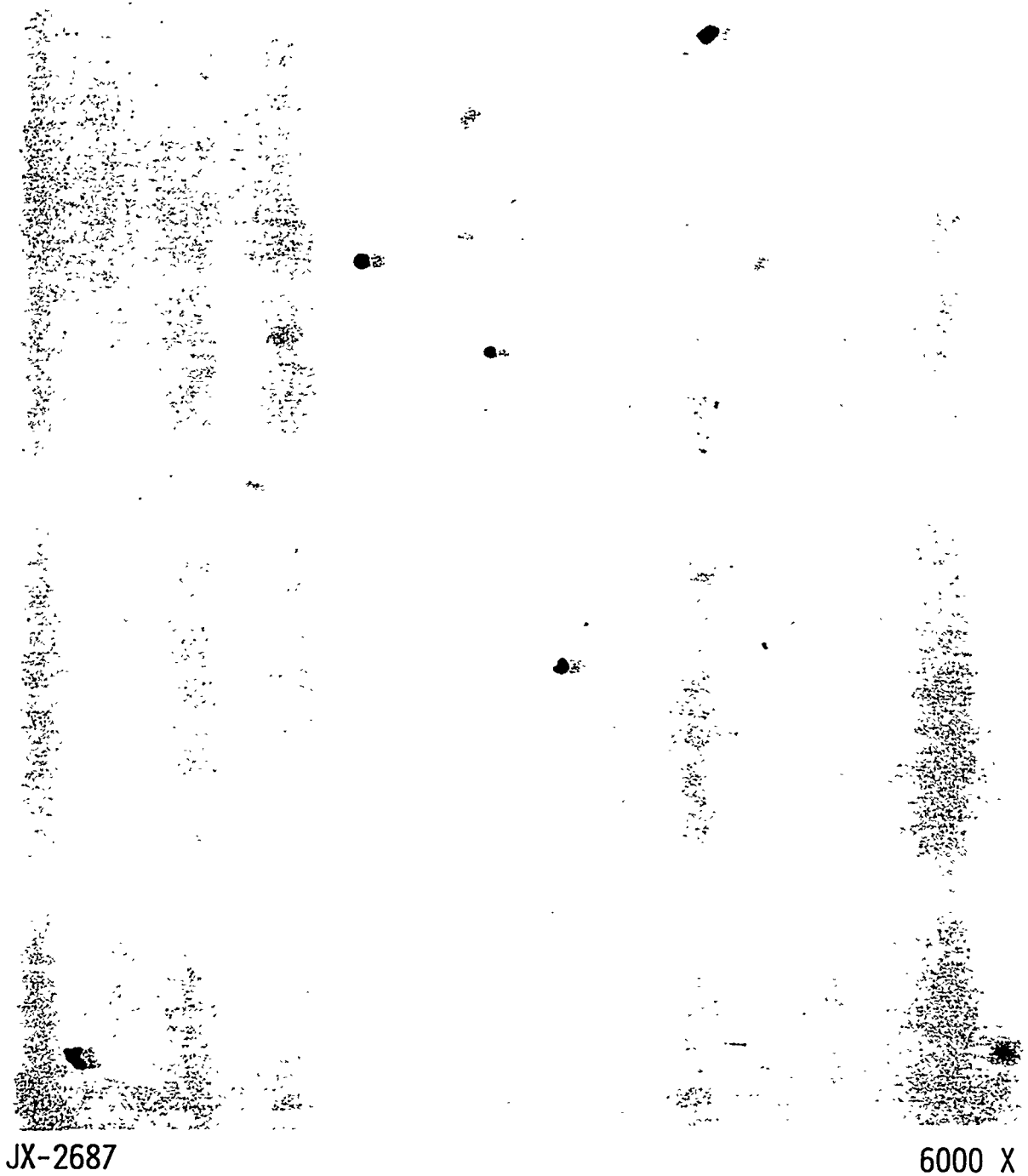


FIGURE 2
AEROSOL PARTICLES COLLECTED FIRST 1½ HOUR
FIRST 1-GRAM RELEASE

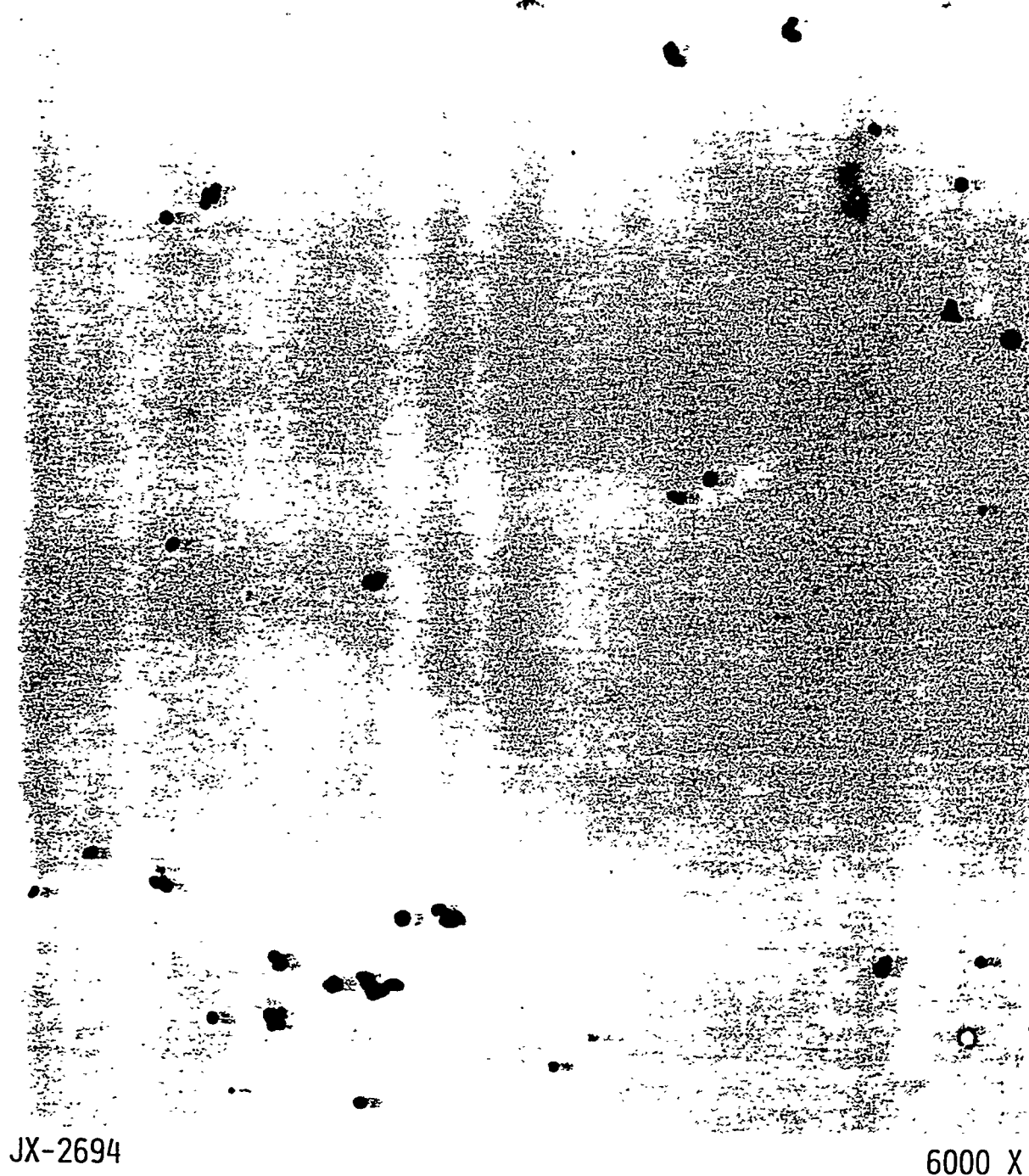


FIGURE 3
AEROSOL PARTICLES COLLECTED FOR 17 HOURS
FIRST 1-GRAM RELEASE

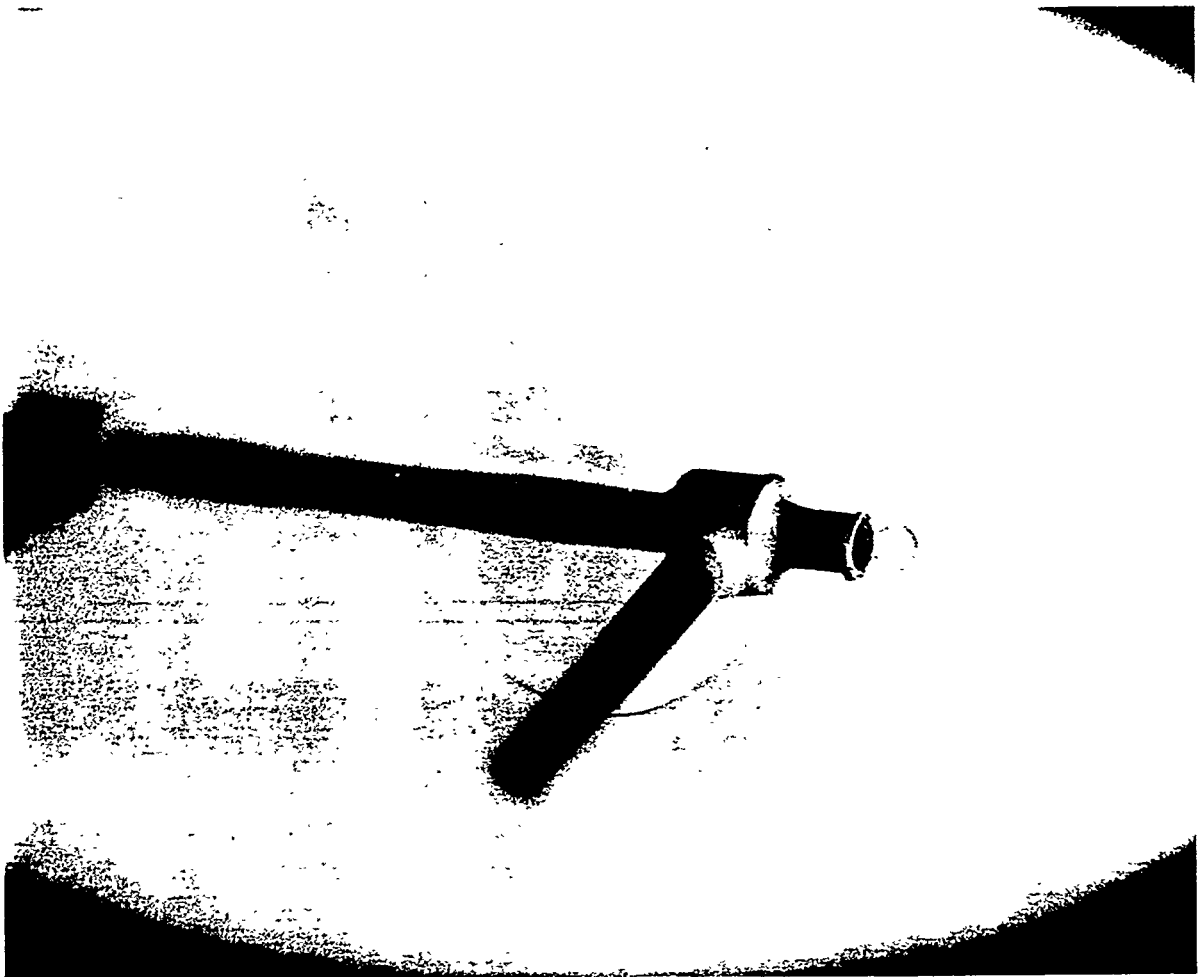
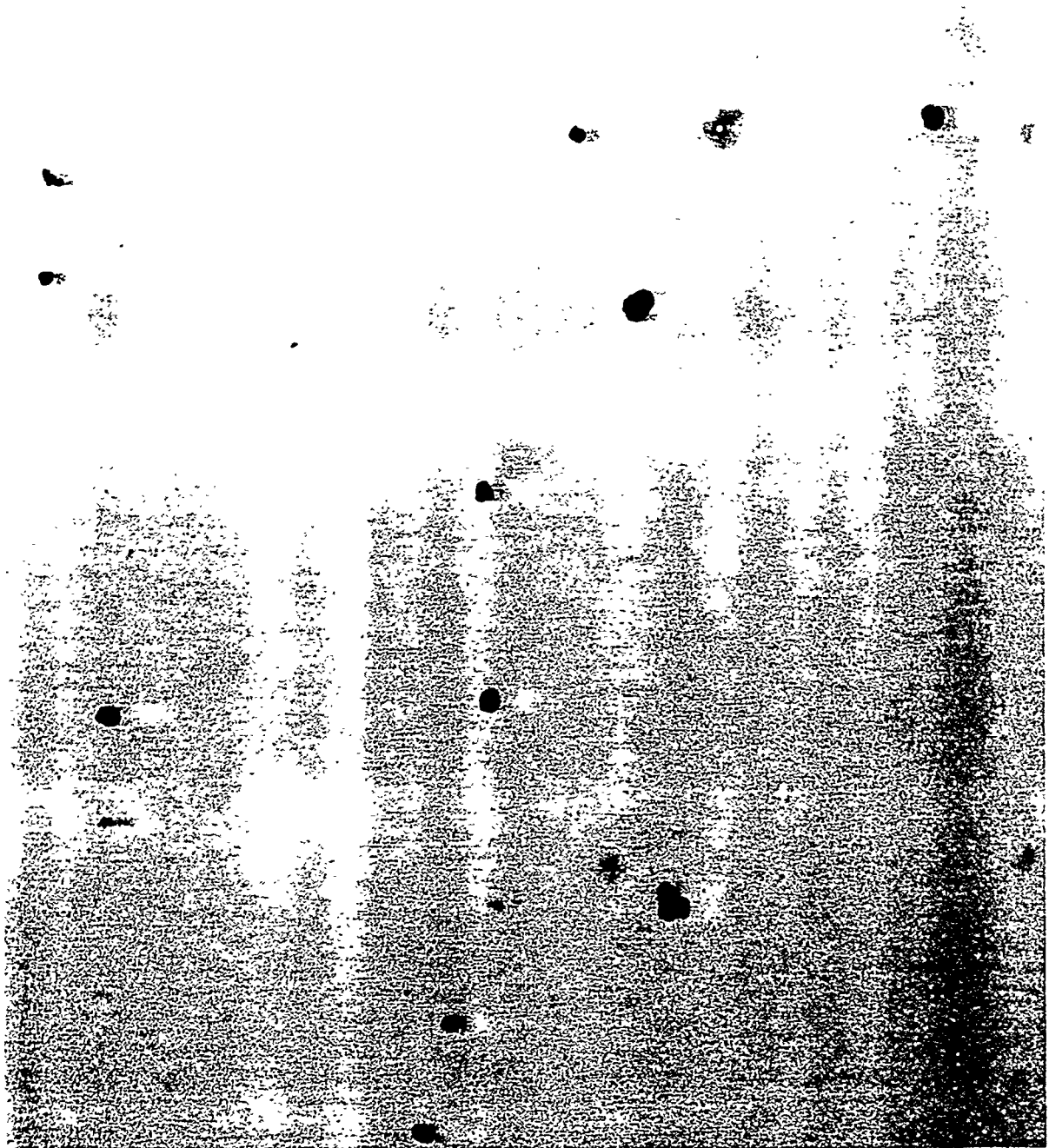


FIGURE 4
PHOTOGRAPH OF CONTAINER AFTER RELEASE
SECOND 1-GRAM RELEASE

Table 1. Aerodynamic particle size distributions of aerosol samples taken at different times after UF_6 release (1 g)

Diameter, Microns	3 Min	17 Min	1/2 h	2 1/2 h	9 h	20 h	27 h	44 h	51 h
18	0	0	0	0	0	7	3	4	0
9	0	0	1	0	0	2	2	2	2
4.5	1	0	0	0	0	0	0	2	0
2.2	2	3	1	2	2	2	2	0	0
1.1	2	2	2	20	41	18	8	2	0
.56	12	20	34	33	25	33	34	24	23
.28	57	43	34	27	17	20	27	31	39
.14	16	19	16	11	9	11	13	19	21
.07	7	8	6	5	4	4	5	12	8
.035	3	5	4	2	2	3	6	4	7

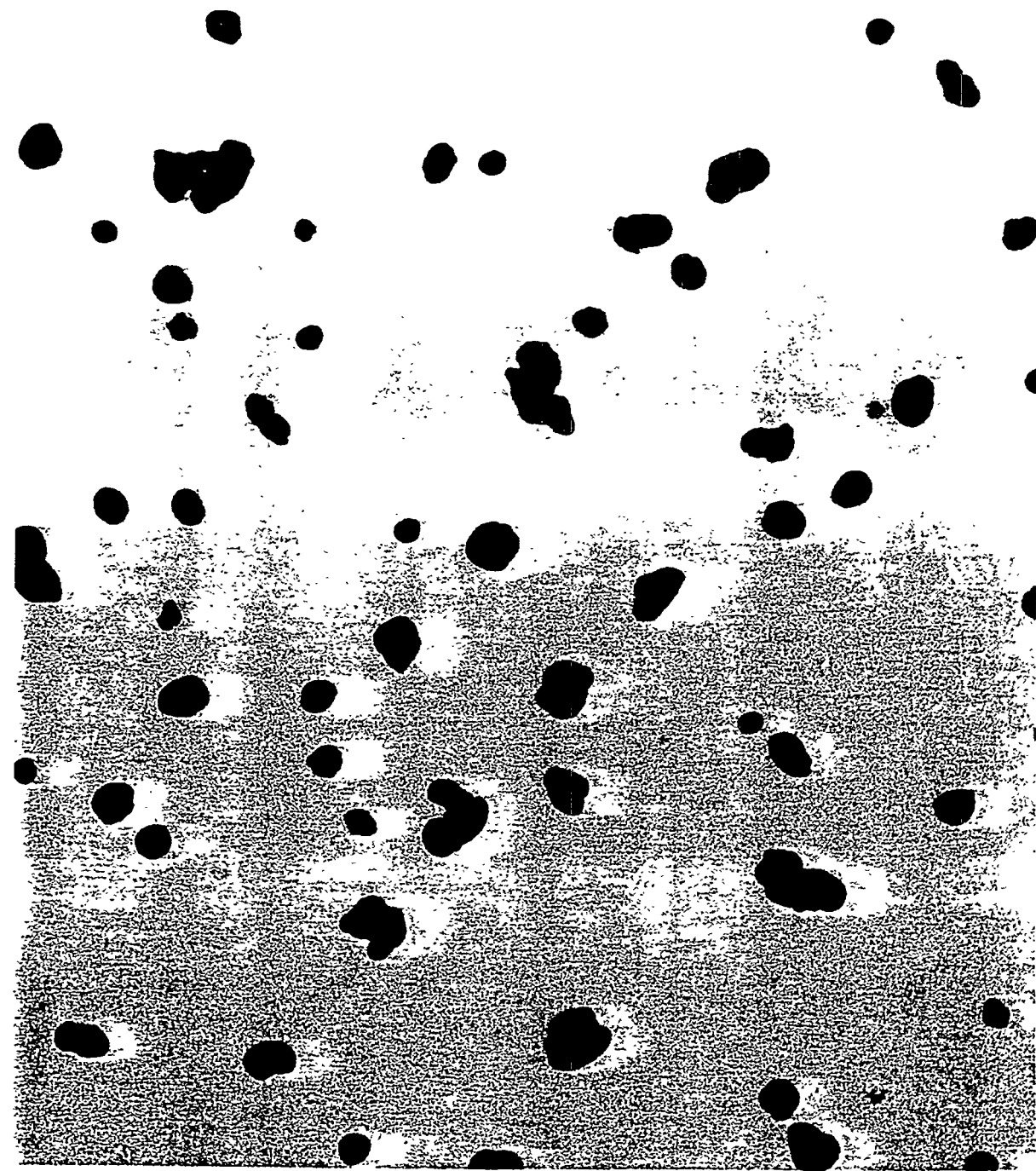
Determination of the mean particle sizes of samples collected for electron microscopy was made by measuring electron micrographs of these samples. Table 2 gives the results of these measurements which follow the size-change pattern observed in the aerodynamic measurements. The diameters were calculated for circles equivalent to the areas of the particles in the micrographs. The last three were collected for 96 h at three different locations on the release chamber floor and show the nonuniform deposition and variation in size of particles caused by turbulence or static electricity in the release chamber. Figures 7, 8, and 9 are electron micrographs which exhibit this segregation by size and number density. Although some "pile-up" of particles (agglomeration after collection) occurred, most of the particles were not altered by the collection process and represent the airborne entities. Figures 10, through 14 are electron micrographs of particles collected for various times and at higher magnification showing the deposit associated with collected particles as they continued to generate hydrolysis product after collection. This feature peaked in samples collected until 1 1/2 h after release and was rarely seen in particles collected after this. An attempt to measure the settling rate of the aerosol particles was not successful because of erratic behavior of the balance being used as a result of turbulence or static electricity in the chamber. Figure 15 is a photograph of the balance pan after settling of the



JX-2843

6000 X

FIGURE 5
AEROSOL PARTICLES COLLECTED FIRST 10 MINUTES AFTER RELEASE
SECOND 1-GRAM RELEASE



JX-2876

6000 X

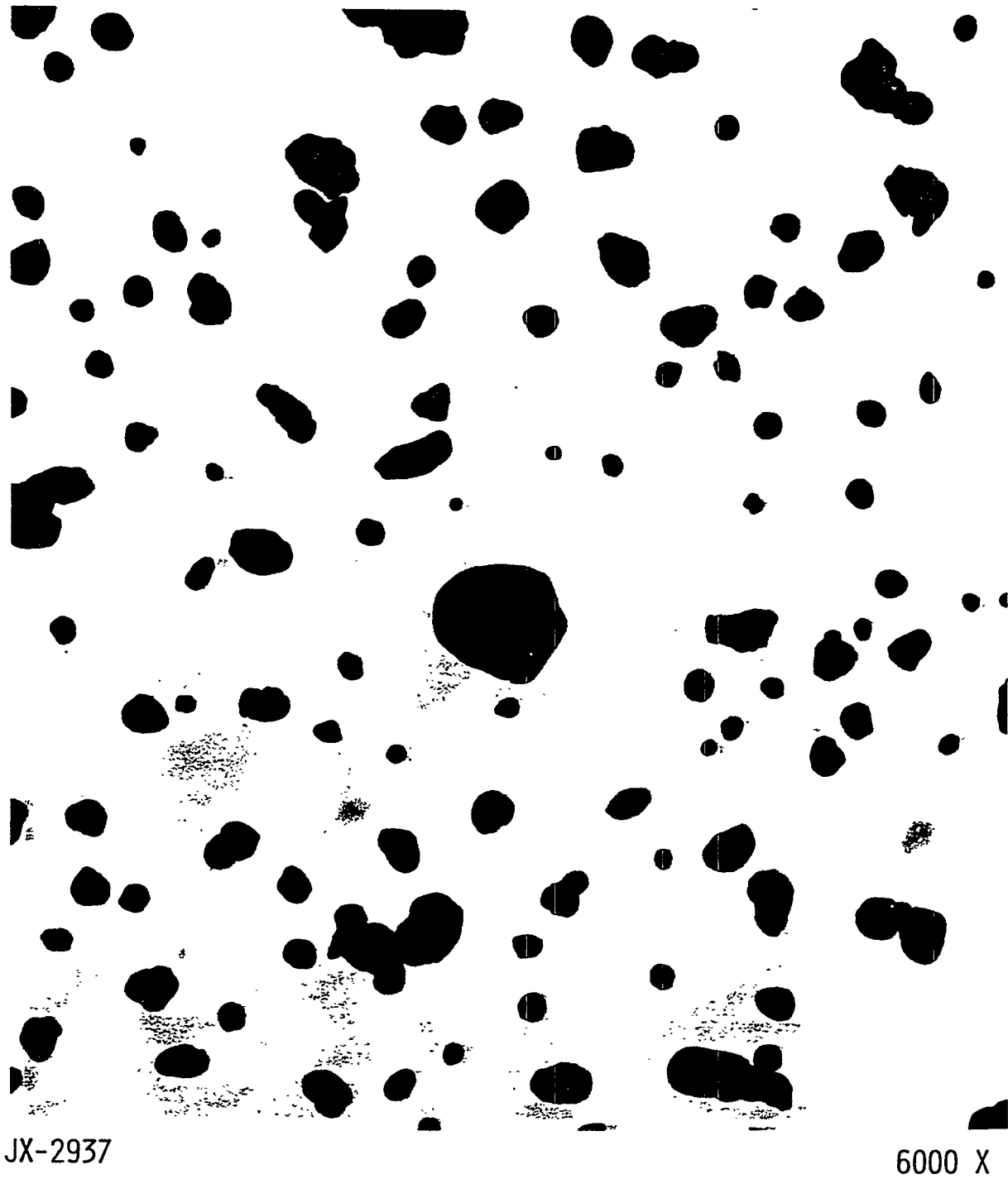
FIGURE 6
AEROSOL PARTICLES COLLECTED FROM 3 HR TO 9 HR AFTER RELEASE
SECOND 1-GRAM RELEASE



JX-2920

6000 X

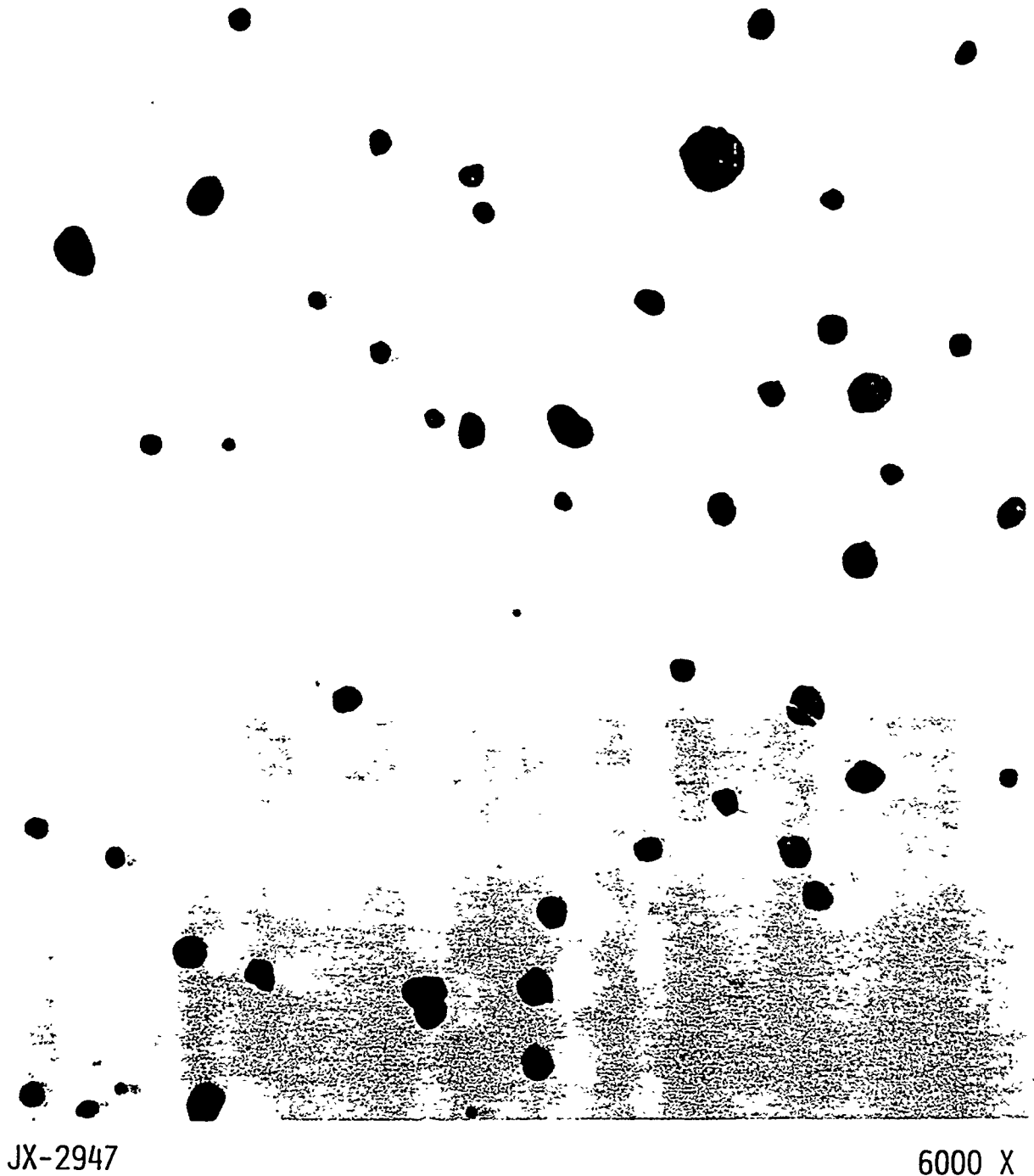
FIGURE 7
AEROSOL PARTICLES COLLECTED FOR 96 HOURS
SECOND 1-GRAM RELEASE



JX-2937

6000 X

FIGURE 8
AEROSOL PARTICLES COLLECTED FOR 96 HOURS
SECOND 1-GRAM RELEASE



JX-2947

6000 X

FIGURE 9
AEROSOL PARTICLES COLLECTED FOR 96 HOURS
SECOND 1-GRAM RELEASE

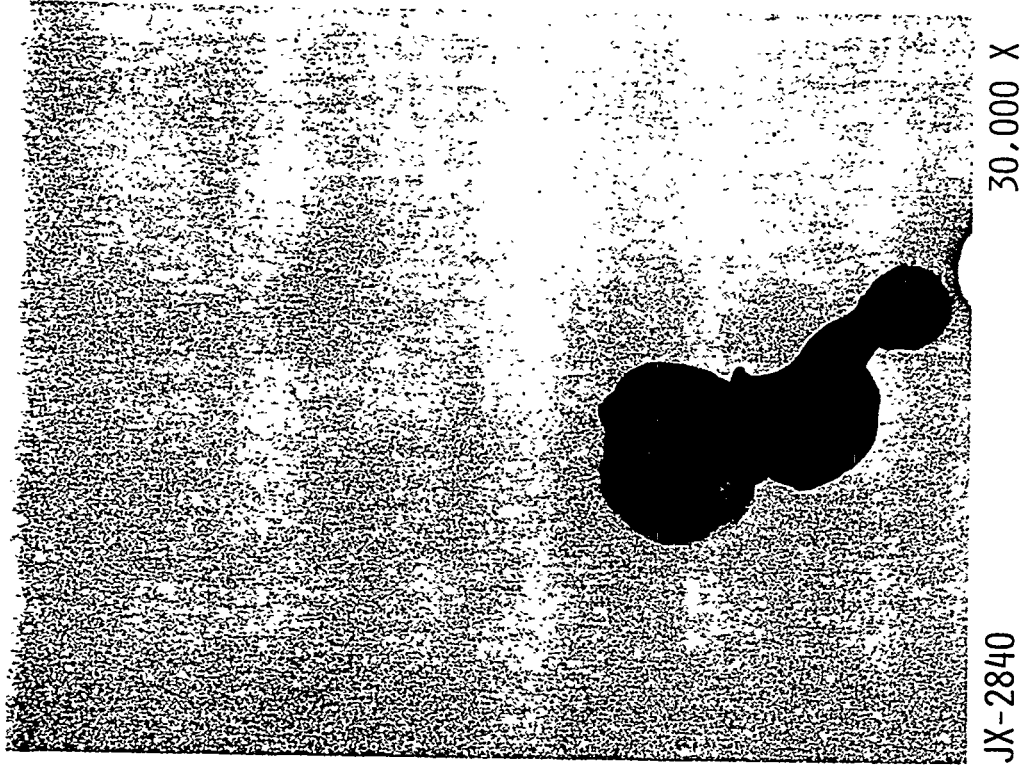
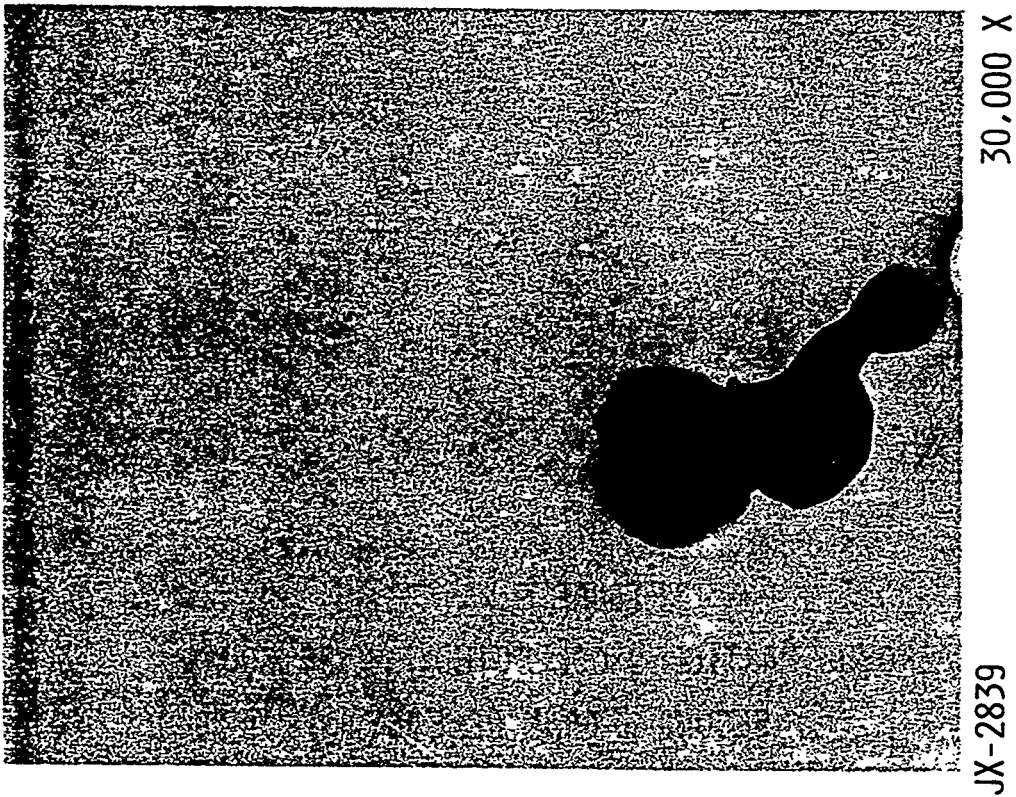


FIGURE 10

AEROSOL PARTICLE FROM SAMPLE COLLECTED FOR FIRST 8 MIN. AFTER RELEASE (STEREO PAIR)
SECOND 1-GRAM RELEASE



JX-2851

30,000 X

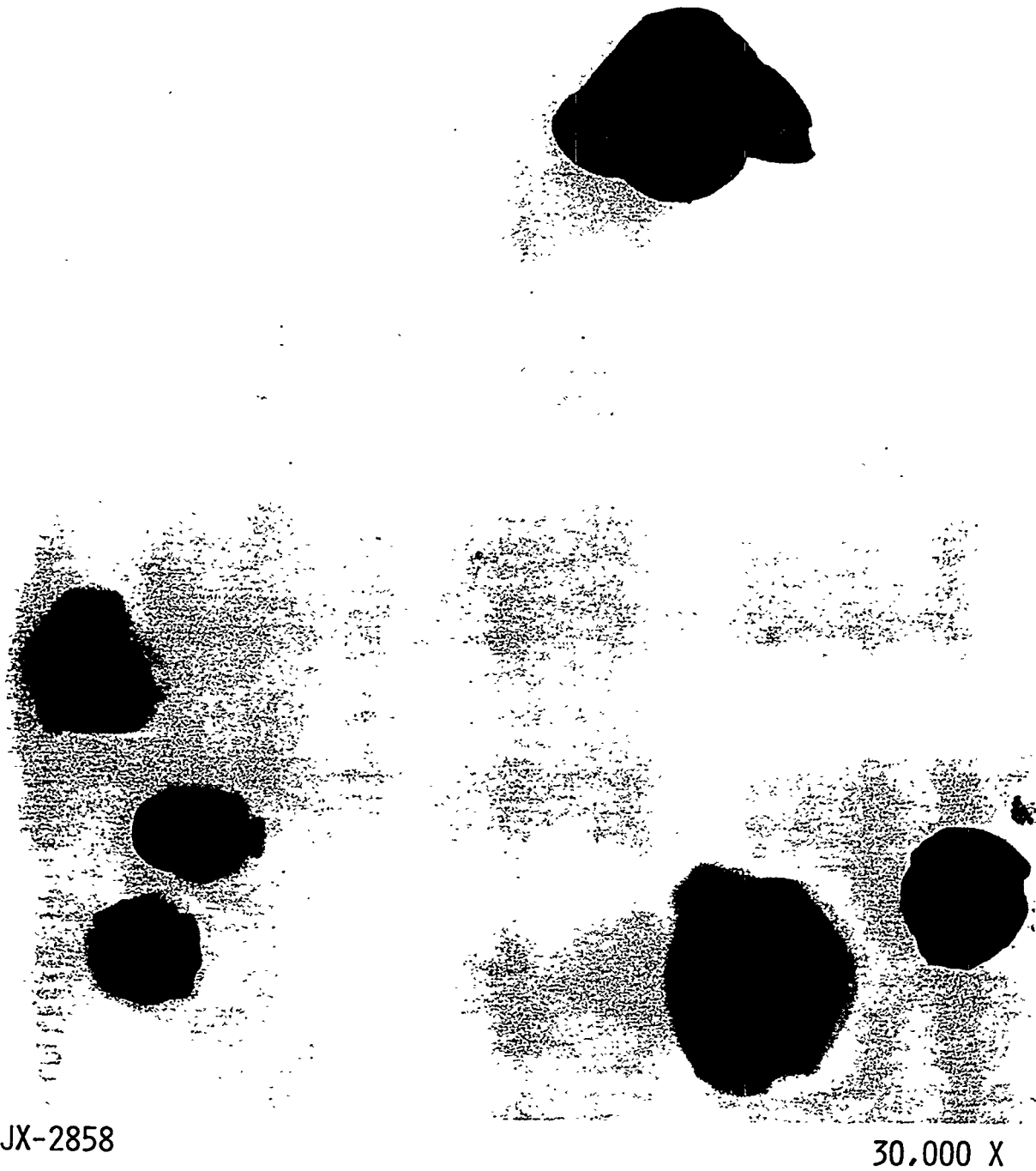


JX-2850

30,000 X

FIGURE 11

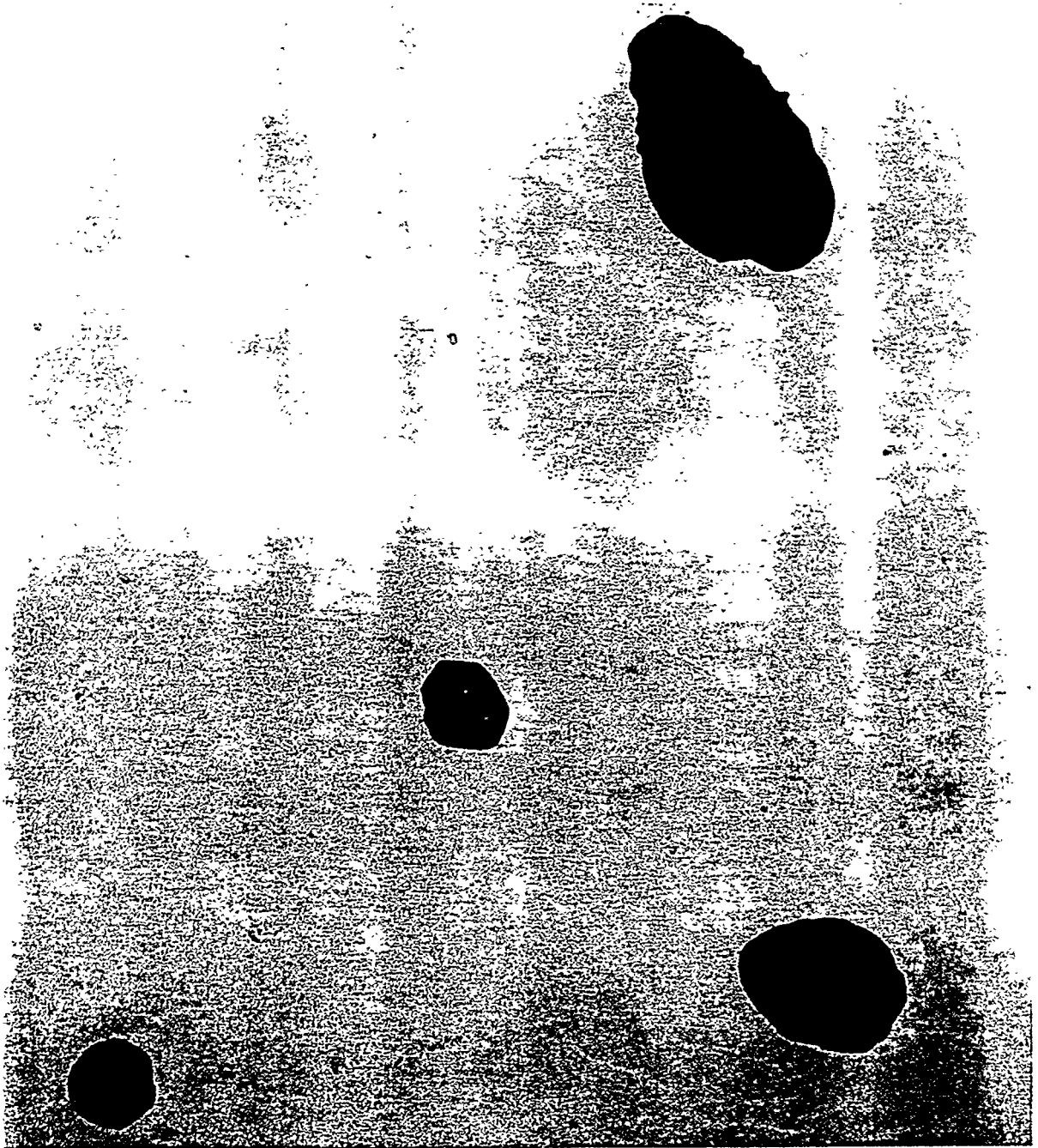
AEROSOL PARTICLES COLLECTED FOR 15 MIN. ENDING 23 MIN. AFTER RELEASE (STEREO PAIR)
SECOND 1-GRAM RELEASE



JX-2858

30,000 X

FIGURE 12
AEROSOL PARTICLES COLLECTED FOR 1 HR
ENDING 1 HR, 23 MIN. AFTER RELEASE
SECOND 1-GRAM RELEASE



JX-2869

30,000 X

FIGURE 13
AEROSOL PARTICLES COLLECTED FOR 70 MIN.
ENDING 2 HR, 33 MIN. AFTER RELEASE
SECOND 1-GRAM RELEASE



JX-2884

30,000 X

FIGURE 14
AEROSOL PARTICLES COLLECTED FOR 6 HR
ENDING 8 HR, 33 MIN. AFTER RELEASE
SECOND 1-GRAM RELEASE

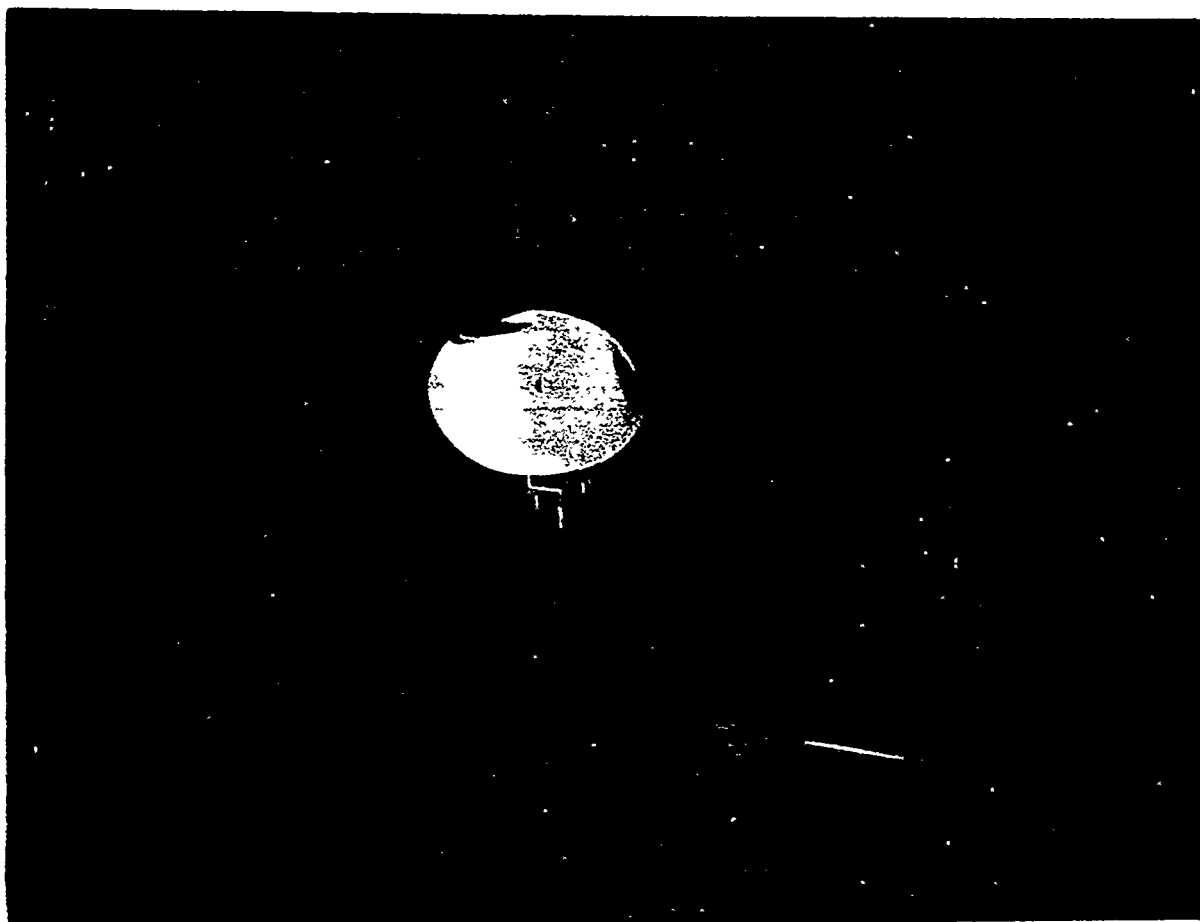


FIGURE 15
PHOTOGRAPH OF BALANCE PAN SHOWING LONG (>1 CM)
AGGLOMERATES OF AEROSOL PARTICLES
SECOND 1-GRAM RELEASE

Table 2. Mean particle diameter variation with elapsed time from electron micrographs

Collection time		Mean Diameter, Micron
Duration	Ending (After release)	
8 min	8 min	.58
15 min	23 min	.66
60 min	1 h, 23 min	.76
70 min	2 h, 33 min	.73
6 h	8 h, 33 min	.97
11 h	19 h, 33 min	.82
7 h	26 h, 33 min	.80
17 h	43 h, 33 min	.70
96 h	96 h	.85
96 h	96 h	.90
96 h	96 h	.76

aerosol showing the relatively long (> 1 cm) agglomerates or stringers of aerosol particles.

ELEVEN-GRAM RELEASE

The release of 11 g of UF_6 was attempted using a metal container with a small valve attached. The release was not as planned since analysis of the container contents after the experiment was concluded showed only 3 g of the 11 g were released because of the restriction and plugging of the valve itself. Even though there was reasonable effort to insure that the valve was kept at a temperature great enough to prevent plugging, the valve construction (it had a polymer plastic core with a relatively small opening) was such that such a temperature could not be maintained. This resulted in a relatively slow release of UF_6 for about 1 min. Figure 16 is a photograph of the container and release plume, the UF_6 not reacting enough to form a visible plume of UO_2F_2 aerosol nearer than a few centimeters from the container in the early stage of the release when the UF_6 pressure was highest. The air in the chamber at time of release was at 20°C and had a relative humidity of 43%.

Electron micrographs show that some of the particles collected exhibit the features attributed to a continuing emanation and hydrolysis of UF_6 . Figures 17 and 18 collected for the first 8 min and the second hour after the release show this effect which persisted to a lesser extent in some of the particles collected for the 1 1/2 h period ending

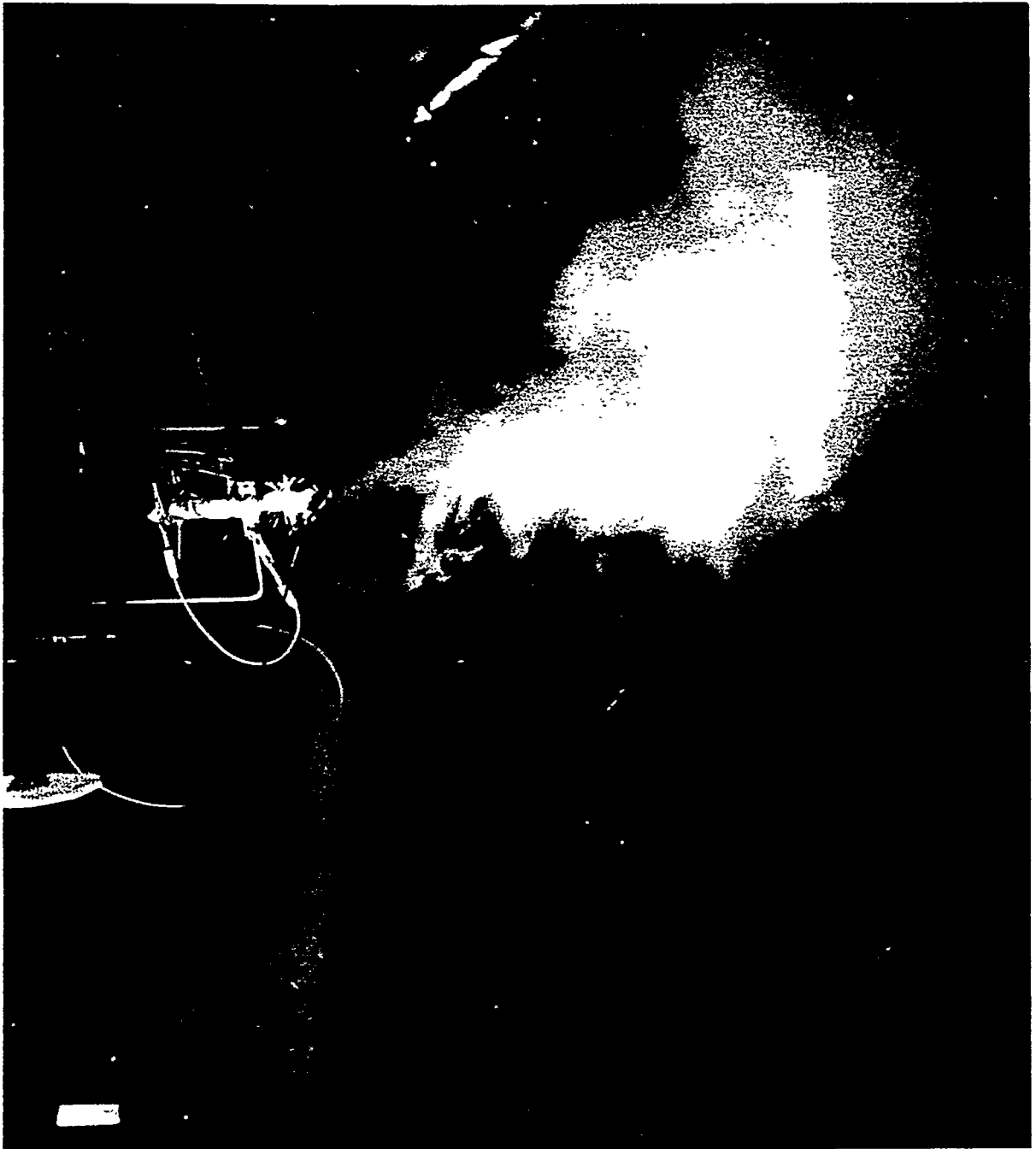
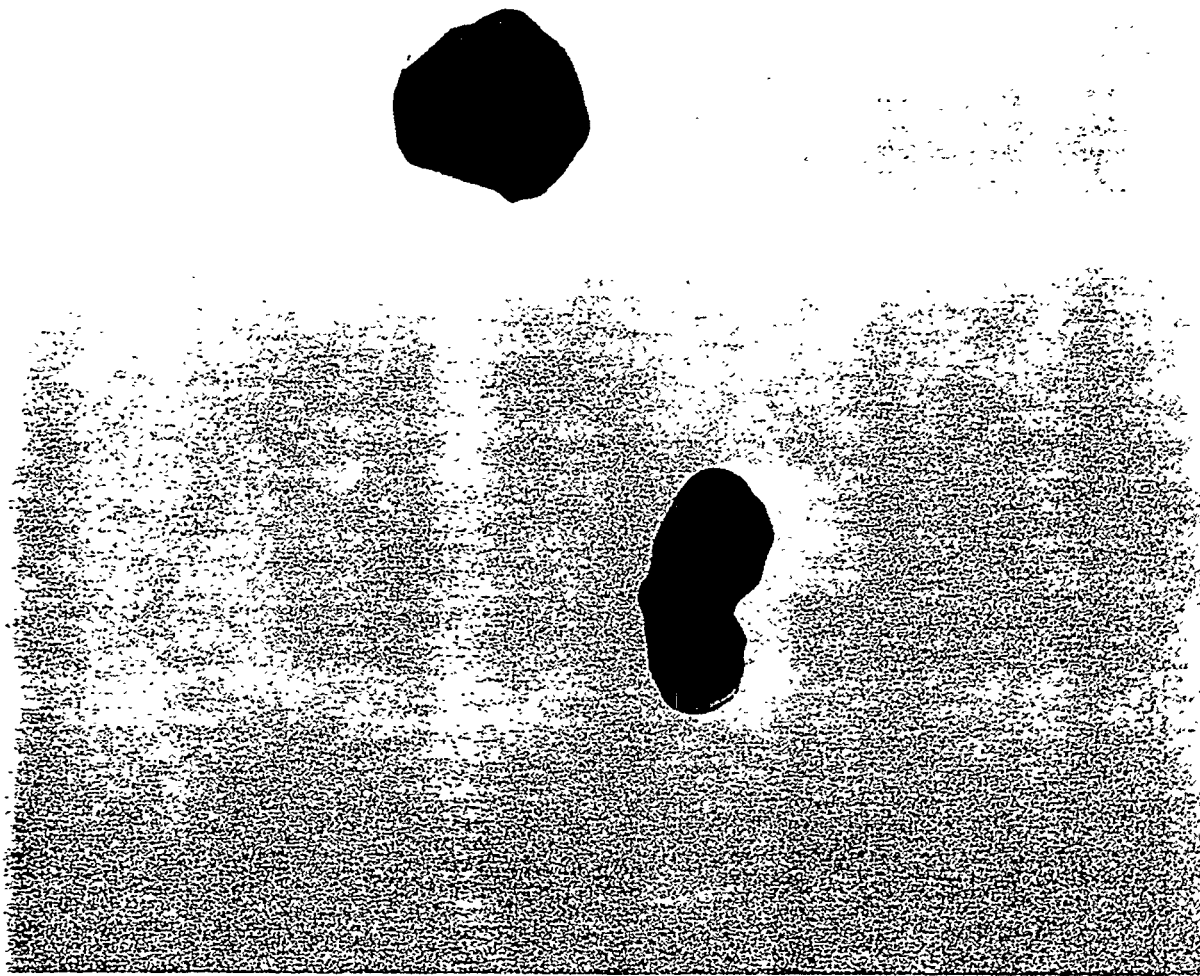


FIGURE 16
PHOTOGRAPH OF RELEASE PLUME
11-GRAM RELEASE



JX-3286

30,000 X

FIGURE 17
AEROSOL PARTICLES COLLECTED FIRST 8 MIN. AFTER RELEASE
11-GRAM RELEASE



JX-3320

30,000 X



JX-3319

30,000 X

FIGURE 18

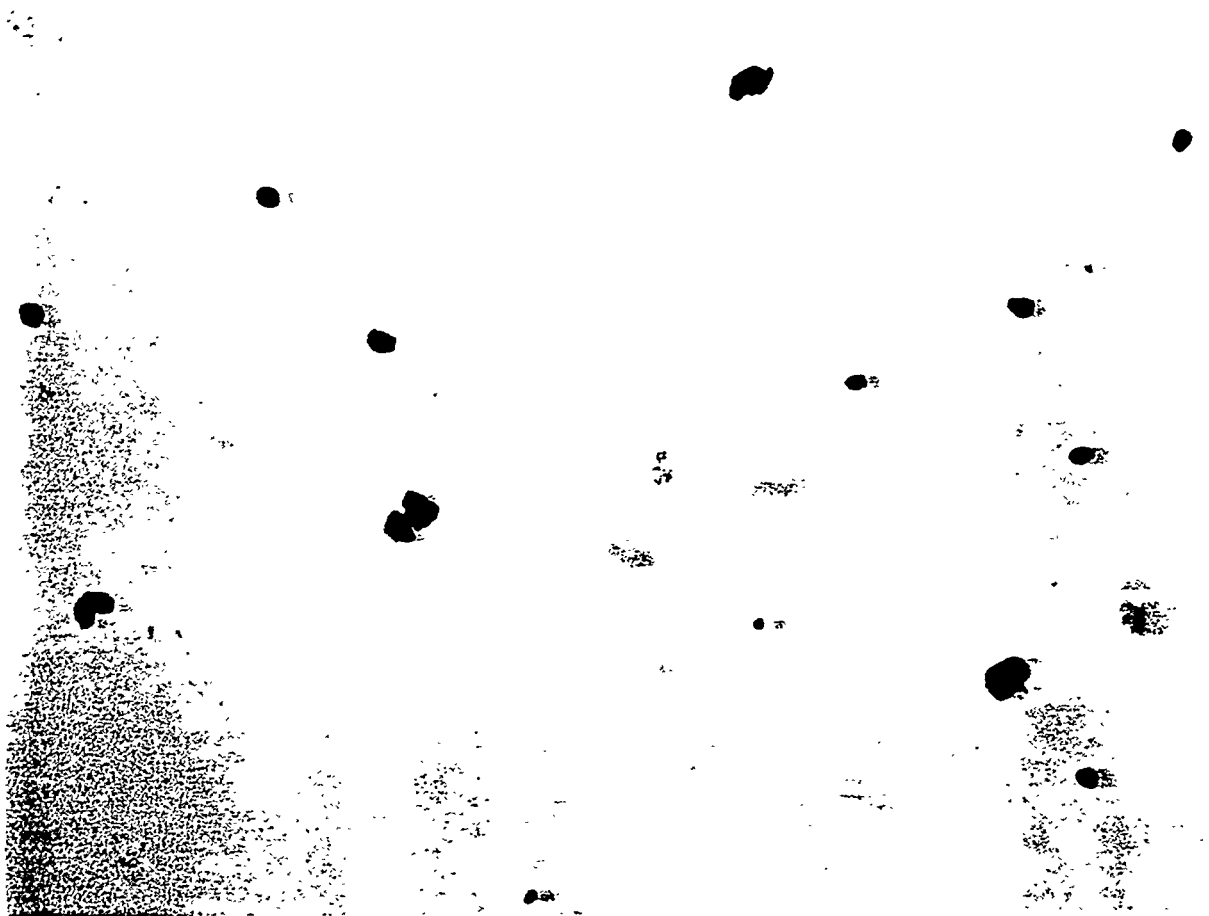
AEROSOL PARTICLE COLLECTED FOR 1 HR ENDING 2 HR AFTER RELEASE (STEREO PAIR)

11-GRAM RELEASE

5 h after the release. Figure 18 also shows a feature quite common in these particles, the apparent sharp shearing or breaking of agglomerates. The reason for this phenomenon is not known. Figures 19 through 22 are electron micrographs which show the spheroidal nature of the particles as they are formed early after release and the tendency to agglomerate soon thereafter. The particles are larger (0.5 to 0.8 μm) than those seen when smaller amounts of UF_6 are released while the agglomerates are very similar in size (up to 10 μm in major dimension) and shape (chainlike) to those seen in smaller releases. Samples collected for longer times (16 h) before fallout decreased aerosol concentration have some larger agglomerates as shown in Fig. 23, but aerodynamic size measurements suggest these are the result of pile-up after collection rather than agglomeration while still airborne. Table 3 tabulates the mean sizes obtained by measuring the micrographs. The sizes quoted were obtained by equating the areas of particles measured to spheres of like areas and since only the particles from the first 8 min sample were approximately equant, the sizes quoted are somewhat artificial. Aerodynamic size distributions obtained at various times after the release are quite typical of other such releases as shown in Table 4 and also suggest the same growth pattern as the micrograph data.

Table 3. Aerosol particle sizes obtained from electron micrographs, calculated mean diameter (11 g)

Collection Time		Mean diameter, Micron
Duration	Ending, after release	
8 min	8 min	.65
13 min	21 min	.89
49 min	70 min	1.1
50 min	2 h	1.3
1 1/2 h	3 1/2 h	1.8
1 1/2 h	5 h	1.6
72 h	100 h	1.3



JX-3293

6000 X

FIGURE 19
AEROSOL PARTICLES COLLECTED FOR 8 MIN. AFTER RELEASE
11-GRAM RELEASE

JX-3295

6000 X

FIGURE 20
AEROSOL PARTICLES COLLECTED FOR 13 MIN.
ENDING 21 MIN. AFTER RELEASE
11-GRAM RELEASE



FIGURE 21
AEROSOL PARTICLES COLLECTED FOR 50 MIN.
ENDING 2 HR AFTER RELEASE
11-GRAM RELEASE

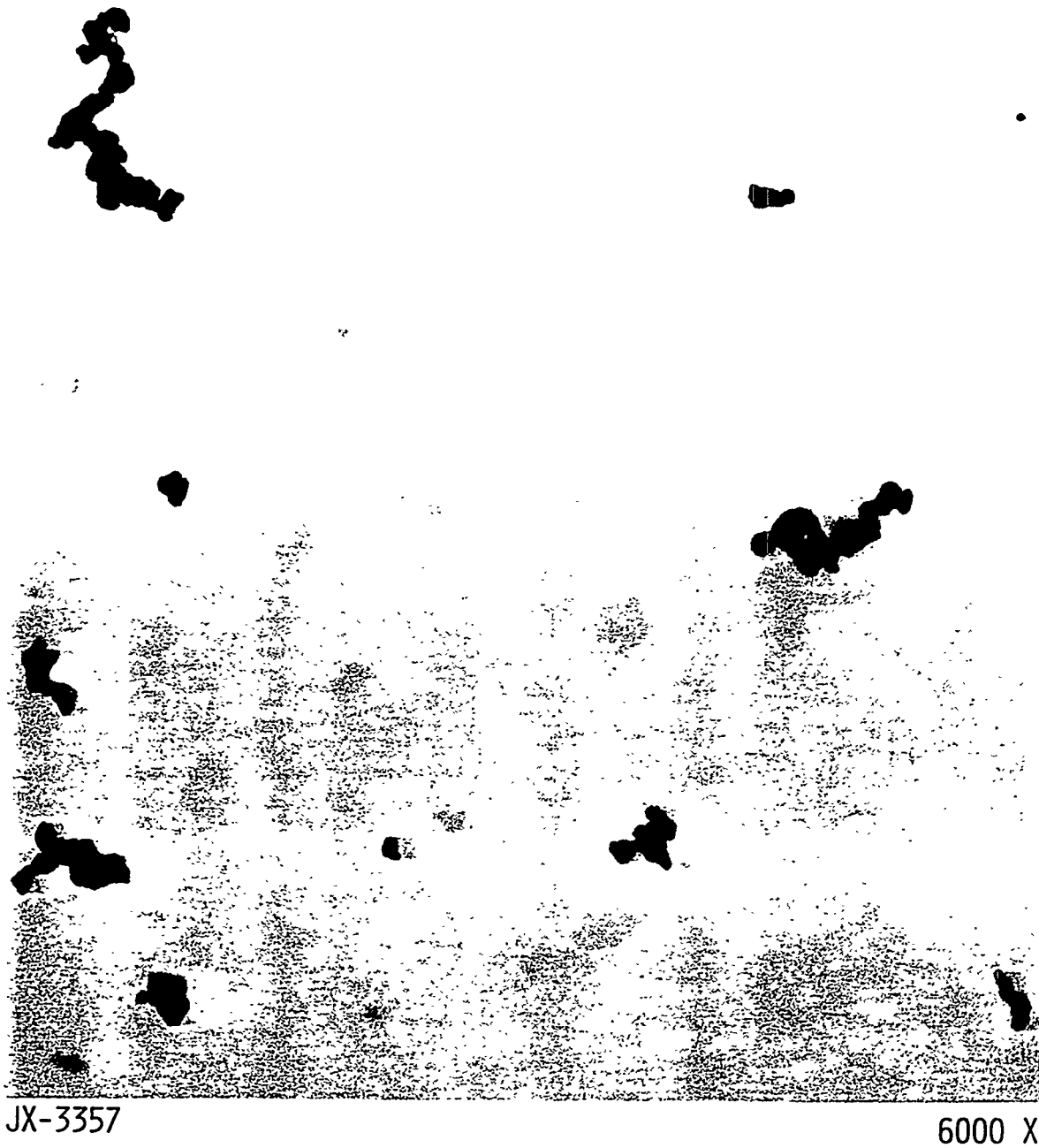


FIGURE 22
AEROSOL PARTICLES COLLECTED FOR 72 HR
ENDING 100 HR AFTER RELEASE
11-GRAM RELEASE



JX-3346

6000 X

FIGURE 23
AEROSOL PARTICLES COLLECTED FOR 16 HR
ENDING 22 HR AFTER RELEASE
11-GRAM RELEASE

Table 4. Aerodynamic particle size distributions of aerosol samples taken at different times after UF_6 release (11 g)

Diameter, μ	Wt % of collected particles at various elapsed times						
	6 min	17 min	1 1/4 h	2 h	3 1/4 h	5 h	6 h
18	0	0	0	0	0	0	0
9	0	0	0	2	0	2	0
4.5	0	0	0	0	2	2	0
2.2	0	0	0	2	1	1	0
1.1	2	2	9	24	32	21	18
0.56	24	24	33	30	27	30	31
0.28	38	40	35	22	17	22	23
0.14	20	17	11	11	11	14	15
0.07	12	11	8	7	8	7	9
0.035	6	6	4	2	2	1	4

THIRTY-FIVE-GRAM RELEASE

The release of 35 g of UF_6 was attempted using a container designed from previous experiences to allow the release to be controlled better, i.e., quickly but not explosively and on demand. Figure 24 is a photograph of a container used subsequently for smaller releases and which has the features incorporated for this release. Instead of the 1/8 in. tubing shown in the figure, 3/8 in. tubing was used to hold the 35 g of UF_6 and Nichrome wire wrapped around the tubing was used as heat source. Release was accomplished by pulling the teflon plug from the O-ring sealed compression fitting. This had the effect of opening the total bore of the tubing, allowing the UF_6 to be expelled without restriction. Release was accomplished in ~ 10 sec and only 5 g of UF_6 was not expelled because of internal condensation and solidification. The UF_6 was at 100°C and the air in the chamber was 21°C and 57% relative humidity at time of release. Figure 25 is a photograph of the floor of the chamber which shows large (visible to the unaided eye) particles of UF_6 which fell to the floor immediately after release and continued to hydrolyze producing the characteristic UO_2F_2 halos. Aerodynamic particle size distributions were obtained on two samples of

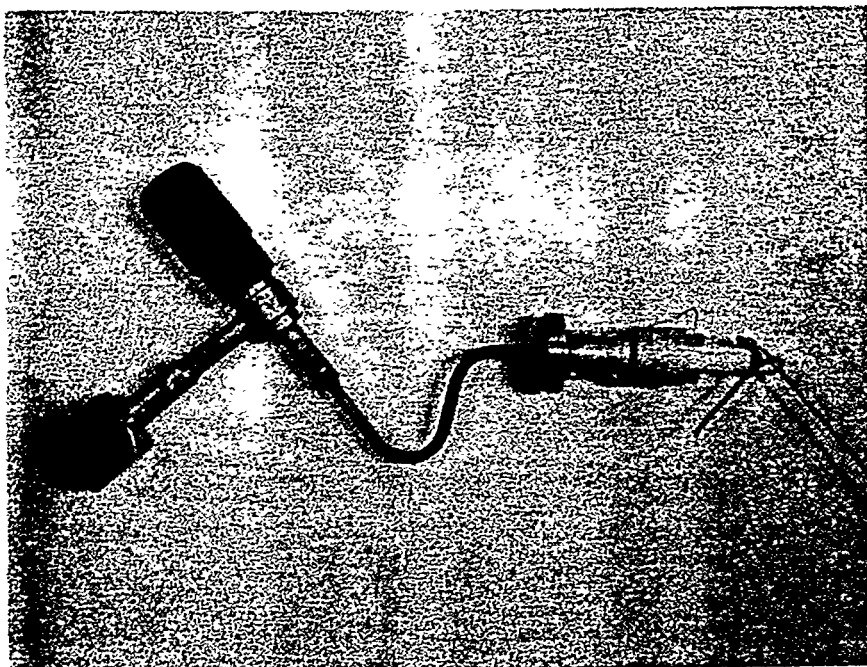


FIGURE 24
PHOTOGRAPH OF CONTAINER USED TO
ALLOW RAPID, CONTROLLABLE UF_6 RELEASE

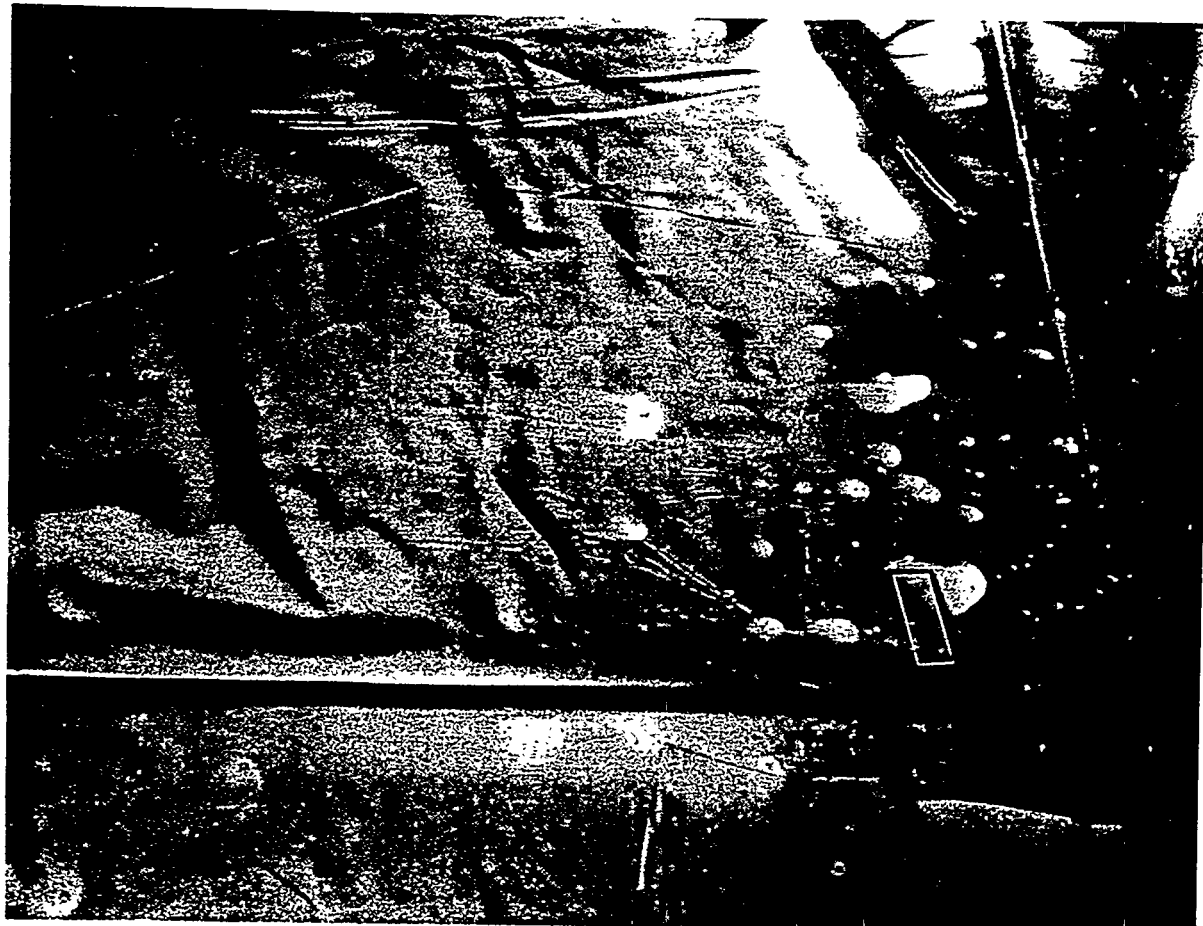


FIGURE 25

PHOTOGRAPH OF FLOOR OF RELEASE CHAMBER AFTER RELEASE
SHOWING LUMPS OF UF_6 AND ASSOCIATED HYDROLYSIS PRODUCT
35-GRAM RELEASE

the aerosol (4 and 25 min after release), but equipment malfunction prevented any further sampling of this type. These two distributions are shown in Table 5.

Table 5. Aerodynamic particle size distributions of aerosol samples from 35 g release

<u>Diameter, μ</u>	<u>Wt % of collected particles</u>	
	<u>4 Min</u>	<u>25 Min</u>
18	0	2
9	0	0
4.5	0	0
2.2	0	1
1.1	1	47
0.56	20	27
0.28	44	9
0.14	25	8
0.07	8	4
0.035	2	2

The particle sizes and the tendency for the particles to grow with time are both typical of lower density aerosols in these distributions. Electron micrographs, however, show the particles collected to be significantly larger and more dense than those from lower density aerosols, especially at longer times. Rather than appearing to be loosely bound agglomerates, the particles are more completely coalesced entities. Particle sizes obtained by measuring electron micrographs are shown in Table 6. Again, these sizes were obtained by calculating diameters of particles from measured areas. Figures 26 through 30 are electron micrographs which show particles collected at various times and which show the growth with time. Fallout of the aerosol was not systematically measured but the rate was not uniform, most of the material settling in the second half hour after release and, unlike less dense aerosols in this chamber, essentially complete settling, as

Table 6. Particle sizes obtained from electron micrographs, calculated mean diameter

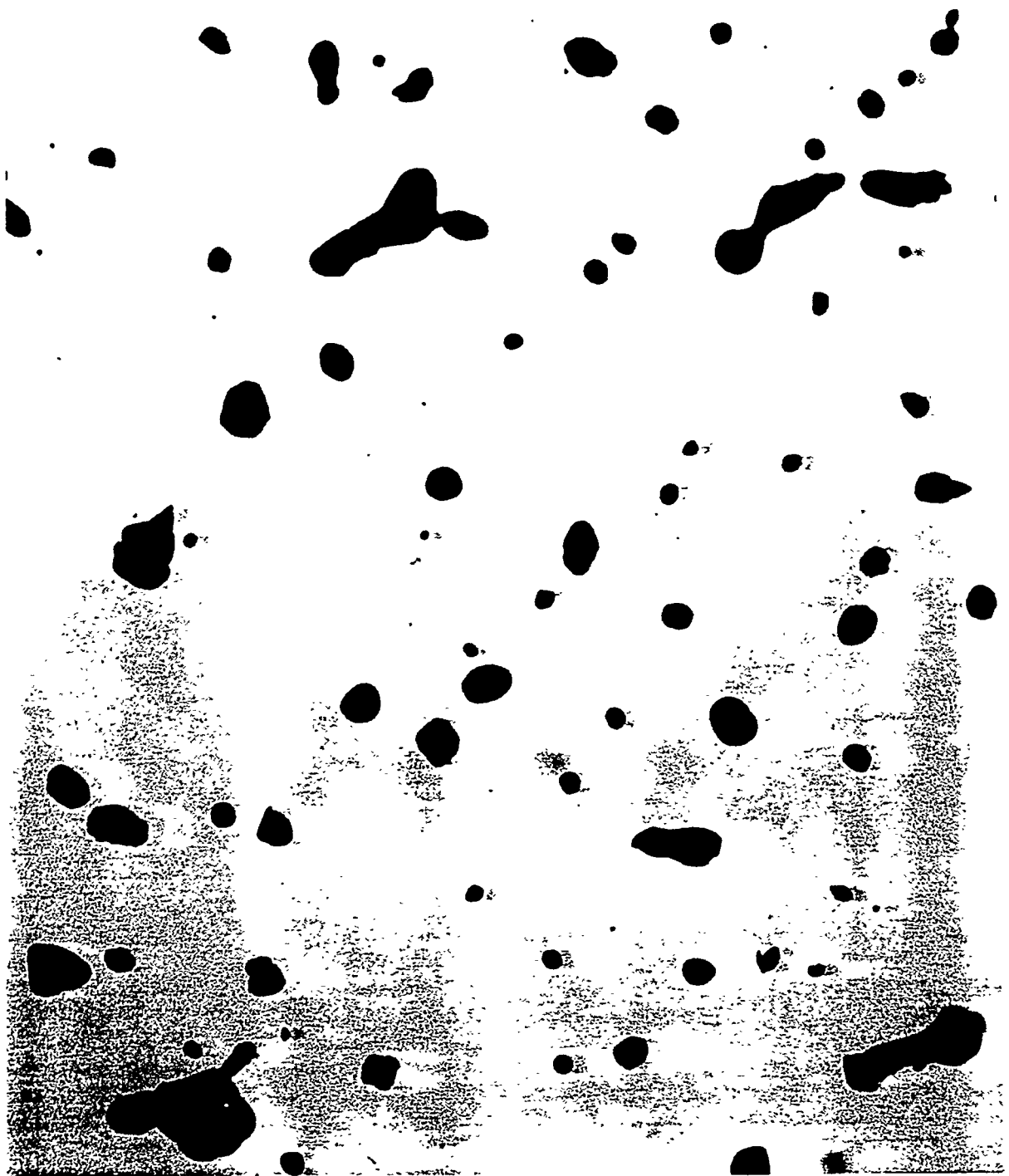
Collection time		Mean Diameter, Micron
<u>Duration</u>	<u>Ending after release</u>	
4 min	4 min	.90
6 min	21 min	2.0
6 min	27 min	2.6
7 min	35 min	3.0
12 h	17 1/2 h	2.0

indicated by laser light scattering, had occurred 24 h after release. Samples collected at intervals from 1/2 to 5 h after release were affected by this settling rate showing too much "pile-up" to be useful. The particles collected for electron microscopy do not have the characteristic secondary hydrolysis halos associated with particles containing unhydrolyzed UF_6 seen in other releases, a surprising and unexplained observation.

X-ray diffraction analysis of material collected from the bottom of the release chamber after complete fallout did not positively identify the phase(s), but the patterns had the general characteristics of uranyl fluoride hydrates or uranyl fluoride hydrate hydrofluorides. Exposure of the sample to ambient air caused changes in the diffraction patterns indicating phase changes were occurring but were still not precisely identifiable. Chemical analysis of the material from the floor of the chamber gave a F/U ratio of > 4 , instead of the expected 2 for $\text{UO}_2\text{F}_2 \cdot \text{XH}_2\text{O}$, indicating HF was present as a hydrofluoride or as an adsorbed species.

ONE HUNDRED FIFTY-GRAM RELEASE

The release of 147 g of UF_6 or 25 g/m^3 was accomplished by using a container designed as that in Fig. 24 but with a 5/8 in. I.D. tube and 1/2 in. diameter O-ring sealed teflon plug. The air in the chamber was at 24°C and 66% relative humidity. The plug was pulled from the container when the UF_6 temperature was 100°C and ~ 25 sec was required for all the UF_6 to be expelled. The release caused much turbulence in the chamber and the aerosol appeared to be well homogenized by the release itself. Weighing of the container after the experiment showed all the UF_6 had been expelled. Uranium content of the aerosol was obtained by analyzing 5 cc samples of aerosol withdrawn at various



JX-3421

6000 X

FIGURE 26

AEROSOL PARTICLES COLLECTED FIRST 4 MIN. AFTER RELEASE
35-GRAM RELEASE



JX-3435

6000 X

FIGURE 27

AEROSOL PARTICLES COLLECTED FOR 6 MIN.
ENDING 21 MIN. AFTER RELEASE
35-GRAM RELEASE



JX-3447

6000 X

FIGURE 28

AEROSOL PARTICLES COLLECTED FOR 6 MIN.
ENDING 27 MIN. AFTER RELEASE
35-GRAM RELEASE



JX-3453

6000 X

FIGURE 29
AEROSOL PARTICLES COLLECTED FOR 7 MIN.
ENDING 35 MIN. AFTER RELEASE
35-GRAM RELEASE

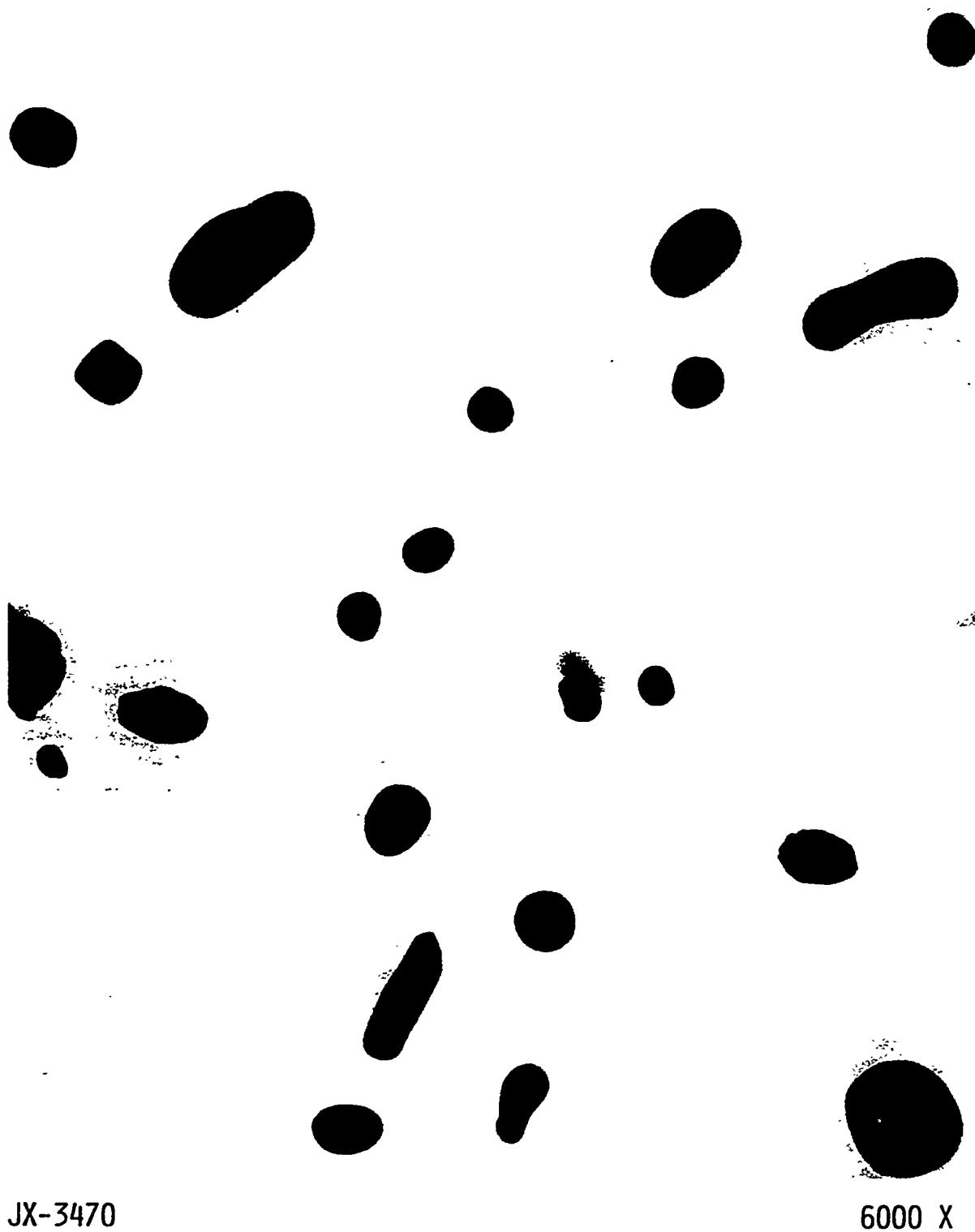


FIGURE 30
AEROSOL PARTICLES COLLECTED FOR 12 HR
ENDING 17½ HR AFTER RELEASE
35-GRAM RELEASE

times. Table 7 gives the results of these analyses and these results are plotted in Fig. 31.

Table 7. Uranium content of aerosol samples withdrawn at various times after release (147 g)

<u>Sample time</u>	<u>U(as UF₆) g/m³</u>
3 min	23.0
7 min	15.8
16 min	9.7
47 min	4.1
101 min	1.84
4 h	1.43

These results show that essentially all of the uranium (UF₆) was airborne, initially, followed by nonlinear fallout. Attempts to get aerodynamic particle size distributions were unsuccessful because of the concentration of the aerosol. Precautions were taken to prevent overloading of the microbalance crystals, but they were inadequate and plugging of the cascaded impactor occurred. Because of the high concentration of the aerosol, electron microscopy samples were collected not by gravitational settling but by centrifugation of the particles from 3 cc of aerosol onto microscopy grids. This method of sampling was used to prevent the obscuring effect of particle "pile-up", but samples collected more than one hour after the release had very few particles. Particles collected soon after the release were much like those collected in other releases, but many particles collected 47 min and later after the release are markedly changed having the characteristics of particles formed in 100% RH experiments. It is not known if the change is real and why or if the change is an artifact caused by the different sampling and treatment technique. Figures 32 and 33 are electron micrographs of particles collected 4 and 17 min after release, respectively, while Fig. 34 is a micrograph of material collected 4 h after release. Particles from a later sample at 5 1/2 h did not exhibit this peculiar morphology, as shown in Fig. 35. Table 8 gives the average sizes obtained by measuring the electron micrographs of three early samples which show the expected increase with time. Later samples were not measured either because of the radical change in particle nature or the lack of particles to measure.

X-ray diffraction analysis of material taken from the chamber 2 h after the release shows the material to be UO₂F₂·1.6H₂O and other

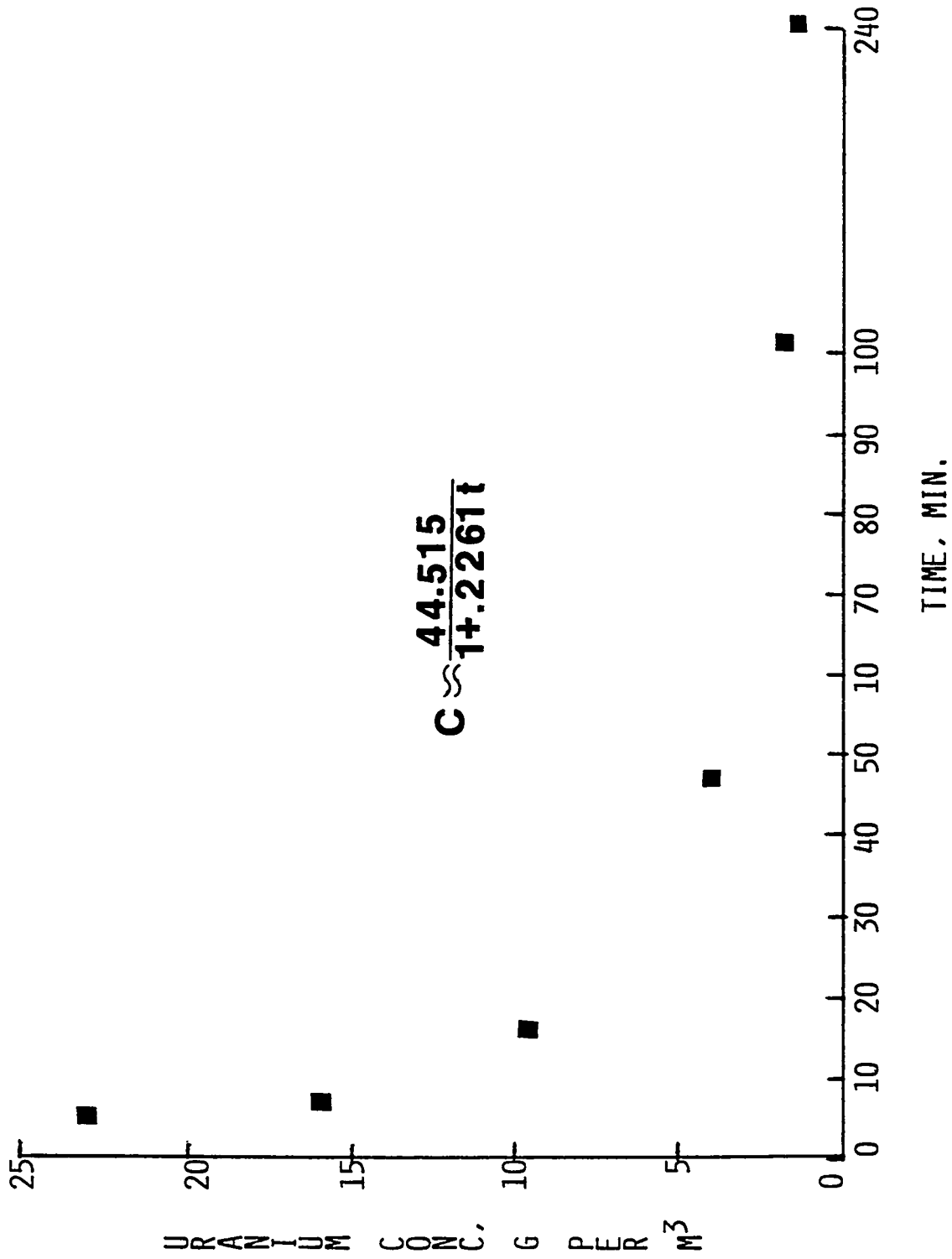
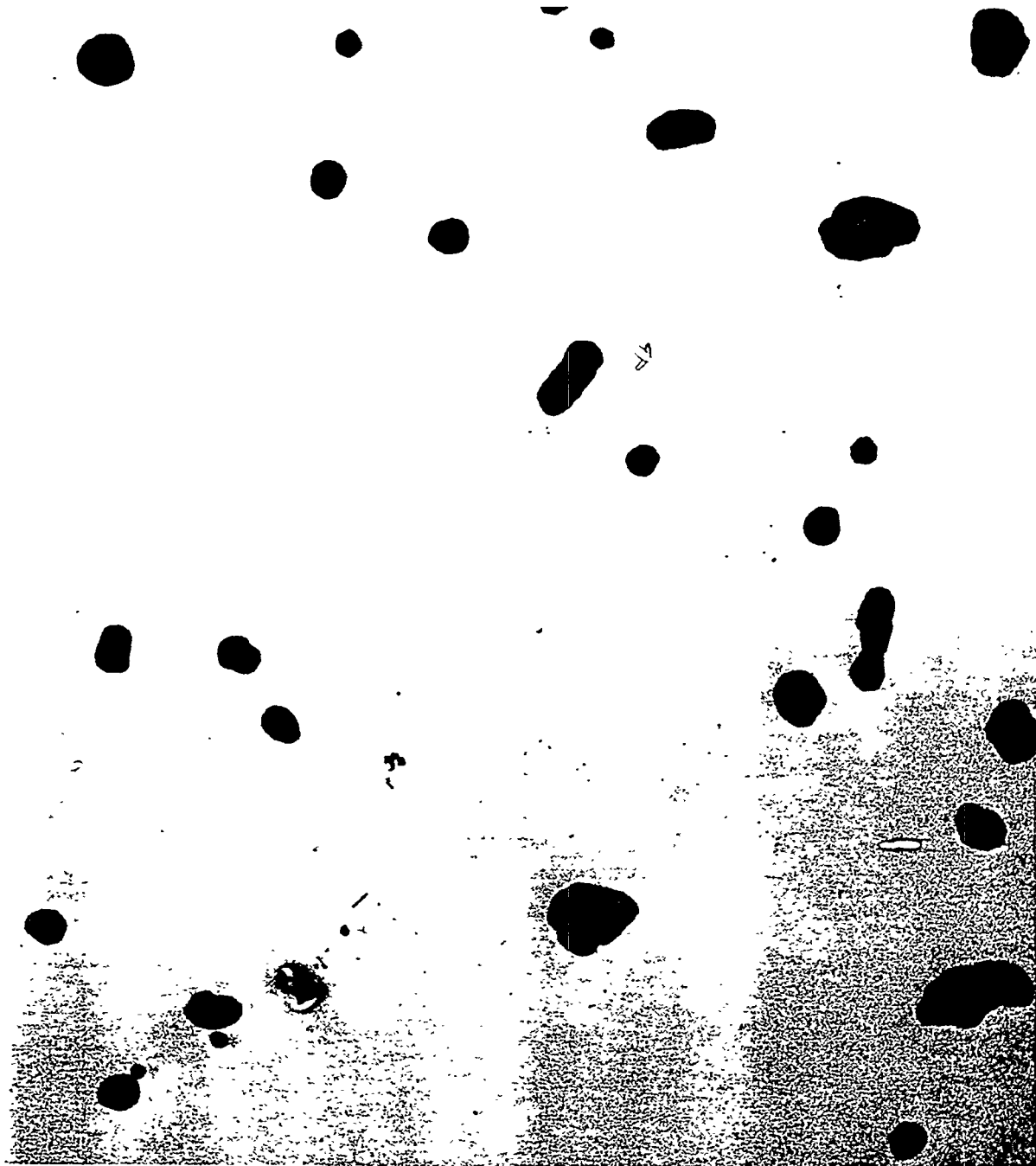


FIGURE 31

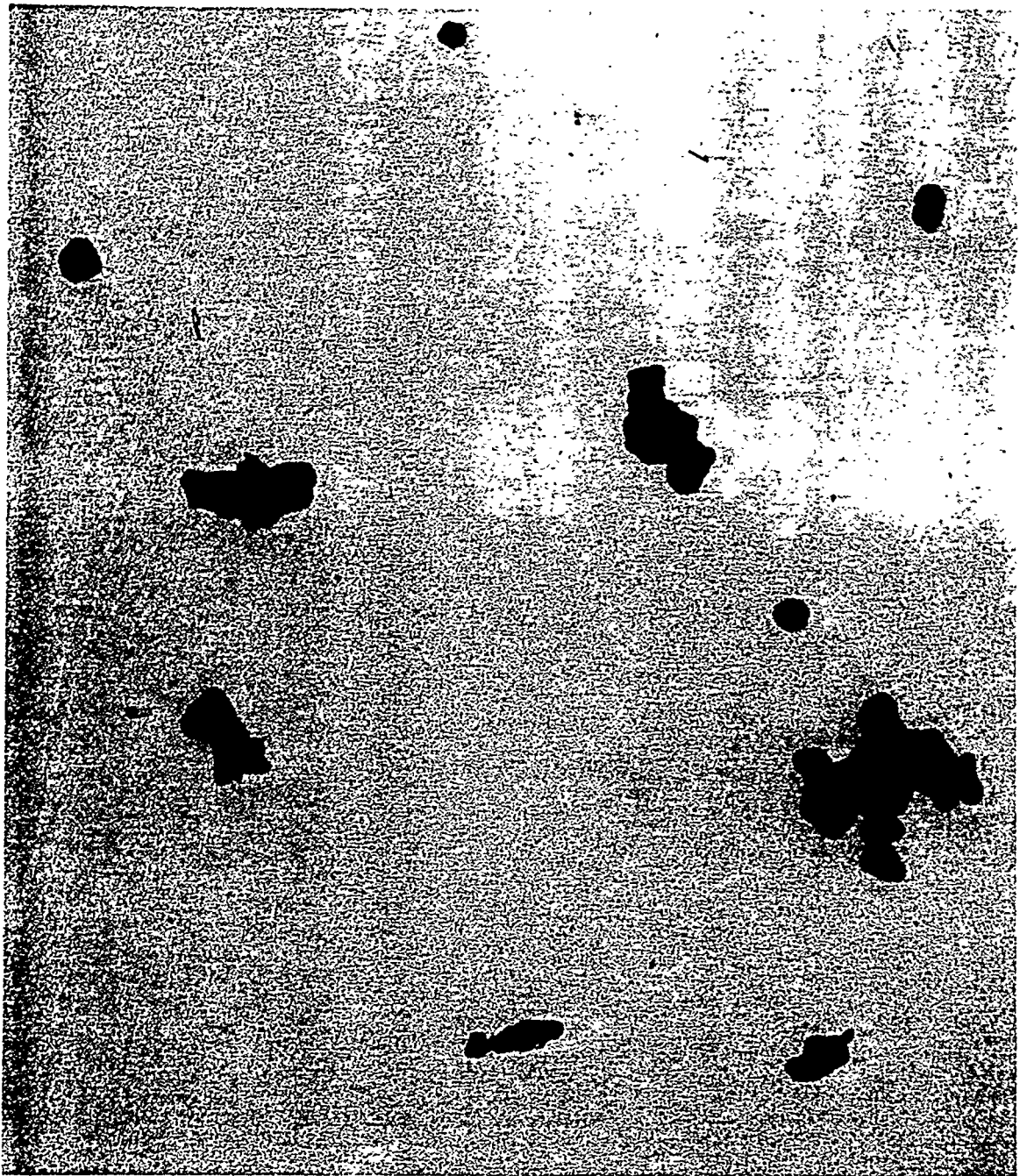
SETTLING RATE OF AEROSOL PRODUCED IN 150-GRAM RELEASE



JX-3732

6000 X

FIGURE 32
AEROSOL PARTICLES COLLECTED 4 MIN. AFTER RELEASE
150-GRAM RELEASE



JX-3756A

6000 X

FIGURE 33

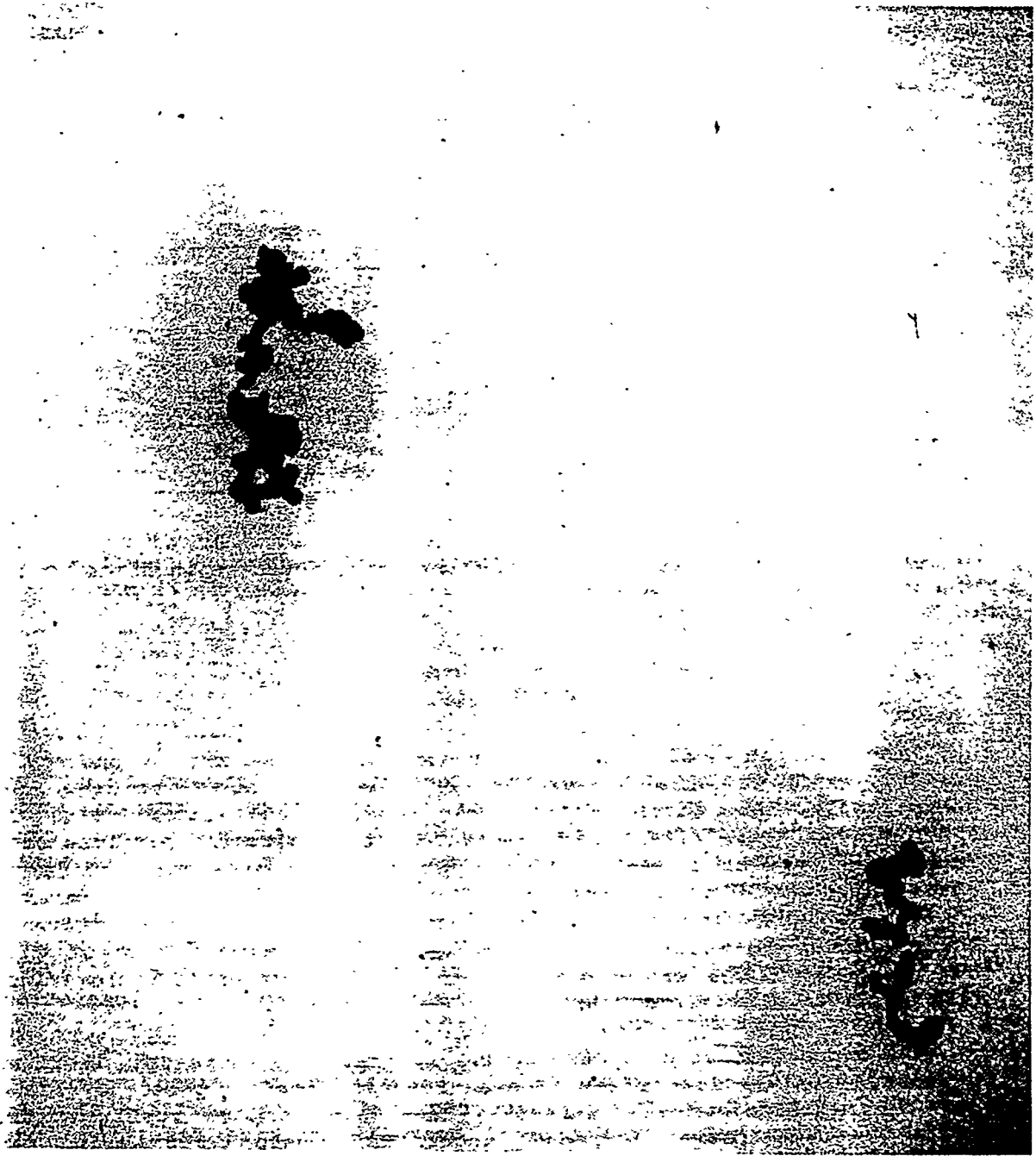
AEROSOL PARTICLES COLLECTED 17 MIN. AFTER RELEASE
150-GRAM RELEASE



JX-3782

30,000 X

FIGURE 34
AEROSOL PARTICLE COLLECTED 4 HR AFTER RELEASE
150-GRAM RELEASE



JX-3788

6000 X

FIGURE 35
AEROSOL PARTICLES COLLECTED 5½ HR AFTER RELEASE
150-GRAM RELEASE

unidentified phase(s), presumably HF adducts from the general nature of the patterns. Samples taken 26 h after release and 2 wk after release

Table 8. Aerosol particle sizes obtained from electron micrographs, calculated mean diameter

<u>Collection time</u>	<u>Mean Diameter, Micron</u>
4 min	1.3
8 min	1.6
17 min	1.9

show only $\text{UO}_2\text{F}_2 \cdot 1.6\text{H}_2\text{O}$. This is consistent with chemical analyses of aerosols in earlier experiments which showed a decrease in HF content of the UO_2F_2 as time progressed.

CONCLUSIONS

The influence of increasing concentration on the morphology of aerosol particles in a static or closed system results, not unexpectedly, in increased particle and/or agglomerate size and sometimes in shape alteration. Measurement of airborne particle sizes from electron micrographs shows the particles to grow and maximize in size at $0.97 \mu\text{m}$ at an initial concentration of 0.17 g/m^3 and $3 \mu\text{m}$ at an initial concentration of 5 g/m^3 . Experimental difficulties prevented measurement of a maximum size at an initial concentration of 25 g/m^3 , but a size of $1.9 \mu\text{m}$ was measured on an early sample while agglomeration or growth was still occurring.

As concentration or amount of UF_6 released was increased, there was a decrease in the time after release required for the particle size to peak or maximize before the typical decrease in size resulting from fallout (concentration decrease) occurred. These times varied from several hours to $\sim 1/2 \text{ h}$.

Aerodynamic size measurements which could be obtained showed the same pattern of initial particle or agglomerate growth followed by a size decrease, but the maximum size of any appreciable fraction of particles was smaller, being $1.1 \mu\text{m}$.

Airborne particles or agglomerates from the lower concentrations had major dimensions of up to $10 \mu\text{m}$, but particles this large were always very nonequant long chains of smaller units. The 5 g/m^3 aerosol

had airborne particles as large as 20 μm in major dimension which were more equant (greater minor dimension) than those seen at lower concentrations.

Some particles appeared to contain unhydrolyzed UF_6 which continued to produce small amounts of UO_2F_2 for some time, as much as 2 h, after formation. Such particles in an outside release would continue to produce smaller, more dispersible UO_2F_2 but probably an insignificant amount.

X-ray diffraction and chemical analyses show the hydrolysis product to be unstable in its behavior, releasing the HF produced by hydrolysis (much of which is associated with the UO_2F_2) and undergoing crystalline phase changes, probably by water absorption and desorption, as well.

These experiments suggest that much of the aerosol produced by UF_6 releases, if given opportunity to disperse or disassociate itself from the release site, will not be readily inclined to "fallout" of and by itself but will require a third body (water droplet, other "dust" particles, etc.) to inhomogeneously be deposited from the atmosphere.

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